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Methods for enhanced delivery of *in situ* remediation amendments in contaminated clay till

Camilla Maymann Christiansen

DTU Environment
Department of Environmental Engineering

PhD Thesis
May 2010

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The thesis will be available as a pdf-file for downloading from the homepage of the department: www.env.dtu.dk

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Preface

The work reported in this PhD thesis, entitled 'Methods for enhanced delivery of *in situ* remediation amendments in contaminated clay till', was conducted at the Department of Environmental Engineering at the Technical University of Denmark with Professor Poul L. Bjerg as supervisor. The PhD project ran from March 2006 to March 2010 and was funded by the Technical University of Denmark and the Capital Region of Denmark. The content of the PhD thesis is based on four papers prepared for scientific journals. The papers represent the sub-projects which were included in the PhD project and conducted in collaboration with internal and external partners. In the text, the papers are referred to by the names of the authors and their appendix number written with roman numbers.

I Christiansen, C.M., Riis, C., Christensen, S.B., Broholm, M.M., Christensen, A.G., Klint, K.E.S., Wood, J.S.A., Bauer-Gottwein, P., and Bjerg, P.L. (2008): Characterization and Quantification of Pneumatic Fracturing Effects at a Clay Till Site. *Environmental Science & Technology* 42 (2): 470-576. DOI: 10.1021/es071294s.

II Christiansen, C.M., Wood, J.S.A., and Bjerg, P.L. (2010): Review of effects of environmental fracturing methods in low-permeability clay-type settings. Submitted manuscript.

III Christiansen, C.M., Damgaard, I., Broholm, M.M., Kessler, T., and Bjerg, P.L. (2010): Direct-Push Delivery of Amendment-Comparable Tracers in Clay Till. Submitted manuscript.

IV Christiansen, C.M., Damgaard, I., Broholm, M.M., Kessler, T., Nilsson, B., Klint, K.E.S., and Bjerg, P.L. (2010): Comparison of Delivery Methods for Enhanced *In Situ* Remediation in Clay Till. Submitted manuscript.

The papers are not included in this www-version, but can be obtained from the Library at DTU Environment. Contact library@env.dtu.dk or Department of Environmental Engineering, Technical University of Denmark, Miljoevej, Building 113, DK-2000 Kgs. Lyngby, Denmark.

Kgs. Lyngby, March 2010
Camilla Maymann Christiansen

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Otto Mønsteds Fond, *Idella*, and *Overlæge Poul Martin Christiansen og hustrus fond* are acknowledged for funding my conference and research trips to Poland and the United States.

Thanks also to collaborators and sparring partners inland and abroad. *Charlotte Riis* and *Anders G. Christensen* (NIRAS) got the ball rolling for a pneumatic fracturing pilot test at a very muddy, cold and windy field site in Vasby, and, thus, all my subsequent work with enhanced delivery methods. Senior researcher *Knud Erik Klint*'s infectious enthusiasm for natural fractures opened my eyes to the fascinating and highly relevant subject of glacial geology, while Professor *Niels Foged*, Associate Professor *Lawrence Murdoch* (Clemson University, SC, USA) and fracturing specialist *William Slack* (FRx Inc.) furthered my understanding of fracturing and related soil mechanics.

Technicians *Mona Refstrup* and *Jens Schaarup Sørensen*, who were unlucky enough to get involved with my field and lab work, are also owed massive thanks for the long hours that we spent together in the dark, hot, and lovingly named 'Gas Chamber'. *Kresten Andersen* (Orbicon | Leif Hansen) and *Bent Skov* are thanked for their work during field injections at the yet again very muddy, cold, and windy Vasby field site – messy and very entertaining!

Illustrators *Torben Dolin* and *Lisbet Brusendorff* are acknowledged for their brilliant graphic minds that have produced great figures for my papers.

Huge thanks go to my friend and former office mate *Katerina Tsitonaki*, who kept me cheerful on tough days and still reminds me on a regular basis that if she could do it, so can I.

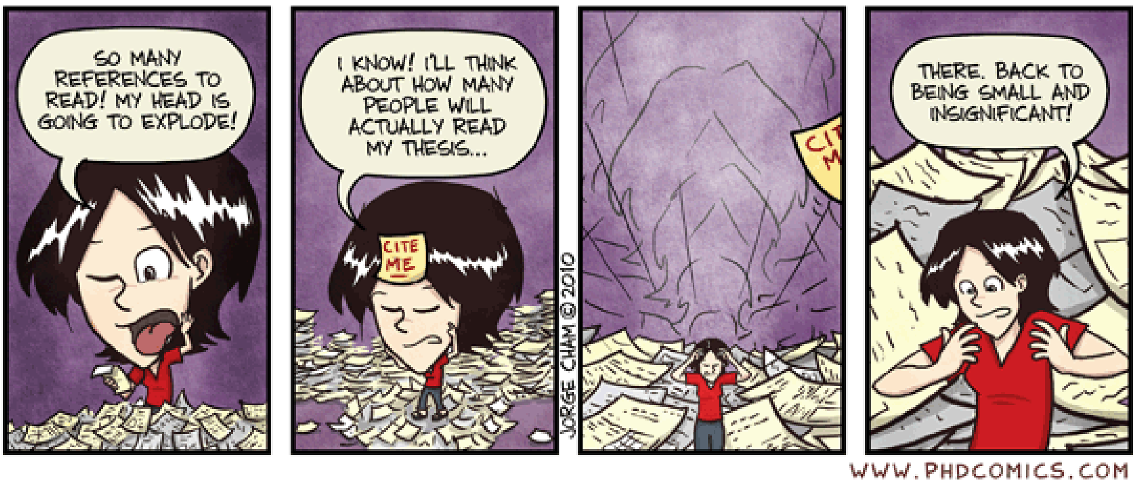
A great big thank you also goes to *Ida Damgaard*, fellow PhD student, friend and office mate, for all her help with the planning and execution of the 2008 pilot tests of hydraulic fracturing and direct-push delivery, and for tying up all the loose ends when I went on maternity leave.

To my other office mates, fellow PhD students and colleagues, thank you for the nice atmosphere – however infrequent a participant I may be, I will miss the cake club, the butter club, and all the other little excuses for a nice chat (and calories).

Luckily, I have close friends and family, who have listened patiently to my complaints and worries during the last four years, and generously offered support and completely un-PhD-related fun and 'hygge' whenever needed. None mentioned, none forgotten, but to *Jude*, *Jane*, and *Julie*: Thanks for being there. And to my mom *Karen*: You are the best!

Very special thanks go to my husband *Jacob* for putting up with me and my mood swings throughout the project. I am eternally grateful to him and my daughter *Signe*, who has kept procrastination at bay during the final writing process.

Last, but not least, I am obliged to thank *Jorge Cham* for "Piled Higher and Deeper", an online PhD comic strip "about life (or lack thereof) in academia"… It has made me laugh on more than one occasion and reminded me that I was not alone…



Summary

Many contaminated sites worldwide constitute a hazard to their surrounding environment and must undergo *in* or *ex situ* remediation. The success of *in situ* remediation depends on achievement of adequate contact between contaminants and amendments in the subsurface. This success criterion is jeopardized in low-permeability deposits, where contaminants have had many years to diffuse into the low-permeability matrix and become virtually inaccessible.

Three technologies may be utilized for enhanced delivery of *in situ* remediation amendments in low-permeability, sedimentary media: pneumatic fracturing, hydraulic fracturing, and direct-push delivery. These methods deliver amendments into the low-permeability deposits by inducing new fractures and utilizing (potentially activating) existing fractures. Induced fracture orientation, form, and radius are dependent on site geology and geotechnical conditions. To facilitate lasting *in situ* remediation enhancements and practicable overall remediation timeframes, closely-spaced (10-25 cm), horizontally-oriented fractures – and hence amendment deliveries – must be achieved.

A thorough literature review has revealed that pneumatic and hydraulic fracturing have been used for the past 20 years without direct documentation of their capabilities at depths exceeding 5 m b.s. Numerous documentation studies have been conducted for hydraulic fracturing at shallow depths (0-5 m b.s.), and published in the peer-reviewed literature. No peer-reviewed studies of pneumatic have been found. Direct-push delivery has only recently been utilized in low-permeability deposits. Peer-reviewed publications on the capabilities of this technology are also lacking.

Pneumatic and hydraulic fracturing have been applied commercially to a large variety of sites. Generally the spacing of emplaced fractures is large (≥1 m) and little or no mention is made of geological and geotechnical site characteristics which could influence fracture propagation. Indirectly documented results (e.g. water sampling) of fracturing-assisted remediation often demonstrate initial enhancement followed by contaminant concentration rebounds, indicating long-term insufficiency of emplaced fractures. In future, those who implement these technologies must, as part of the remedial design, consider what fracture/delivery characteristics are necessary to achieve set remediation goals within a reasonable timeframe, and whether geological and geotechnical site characteristics are amenable to this.

Thus it is clearly necessary to raise awareness, via the scientific literature, of how site geology and geotechnical features influence induced fracture characteristics and, hence, what is achievable with the enhanced delivery methods under various conditions. Contributing to direct documentation of all three methods at depths exceeding 5 m b.s. is important, as it is presently unknown whether the enhanced delivery technologies can fulfill the requirements for effectively enhanced remediation at these depths.

A field study has therefore been conducted, for the first time incorporating testing and direct documentation of the capabilities of all three enhanced delivery methods at depth (2.5-9.5 m b.s.). Direct documentation at depth was largely confined to coring, but was supplemented by excavation at shallow depths. A clay till site in Denmark was selected for the study, as clay till is an abundant low-permeability, sedimentary, and often contaminated deposit in Northern Europe and North America.

The study demonstrated that hydraulic fracturing functioned well at 3 m b.s. However, attempts to emplace horizontal (closely-spaced) fractures at 6-7 and 9.5 m b.s. were unsuccessful. Induced pneumatic fractures (4-8 m b.s.) were initially horizontal, but prone to diversion in natural (vertical) fractures in the sediment. Close networks of fractures at each fracturing depth were not observed. However, discrete, closely-spaced fractures may be obtainable if a smaller spacing is implemented between fracturing intervals. Direct-push delivery was successful in creating closely-spaced, horizontal substance deliveries at all tested depths (2.5-3.5, 6-7, and 8.5-9.5 m b.s.).

These findings correspond well with expectations, given the geological and geotechnical features of the chosen clay till site (a normally consolidated, extensively naturally fractured basal clay till). The findings furthermore emphasize the need for thorough geological characterization and geotechnical testing at all *in situ* remediation sites that could potentially be assisted by enhanced delivery.

Unresolved issues remain. For pneumatic and hydraulic fracturing the main unresolved issues are: 1) the lower limits of fracture spacing at depths greater than 5 m b.s., and 2) the ability of these technologies, especially hydraulic fracturing, to create subhorizontal fractures at depths greater than 5 m b.s. For direct-push delivery, the influence of delivery volumes on delivery orientation, form, and radius is unclear.

Dansk sammenfatning

Mange forurenede lokaliteter verden over udgør en trussel for deres omkringliggende miljø og må oprenses *in* eller *ex situ*. *In situ* oprensningssucces afhænger af tilstrækkelig kontakt mellem forurening og oprensningsmidler i jorden. Indfrielsen af dette succeskriterium vanskeliggøres i lavpermeable sedimenter, hvor forureningen har haft mange år til at diffundere ind i den lavpermeable matrix og blive mere eller mindre utilgængelig.

Tre teknologier kan anvendes til forbedret tilførsel af *in situ* oprensningsmidler i lavpermeable, sedimentære medier: pneumatisk frakturering, hydraulisk frakturering og direkte sonde tilførsel. Metoderne tilfører oprensningsmidler til lavpermeable sedimenter via dannelse af nye sprækker og anvendelse/åbning af eksisterende sprækker. Nydannede sprækkers orientering, form og radius afhænger af lokale geologiske og geotekniske forhold. For at opnå vedvarende oprensningsforbedringer og praktisk gennemførlige oprensningstider skal tætliggende (10-25 cm), horisontalt udbredte sprækker – og dermed stoftilførsler – opnås.

Et dybdegående litteraturstudie har vist at pneumatisk og hydraulisk frakturering har været anvendt gennem de sidste 20 år uden direkte dokumentation af deres evner ved dybder, der overstiger 5 m u.t. Adskillige dokumentationsstudier for hydraulisk frakturering ved lave dybder (<5 m u.t.) er blevet udført og publiceret i den tilgængelige litteratur. Peer-reviewede studier af pneumatisk frakturering er ikke blevet fundet. Direkte sonde tilførsel har kun været anvendt i lavpermeable medier i få år, hvormed tilgængelige publikationer vedrørende denne teknologis evner også mangler.

Pneumatisk og hydraulisk frakturering er blevet anvendt kommercielt på en lang række lokaliter. Generelt er afstanden mellem udførte sprækker stor (≥ 1 m) og få eller ingen overvejelser bliver angivet for geologiske og geotekniske faktorers forventelige indflydelse på sprækkernes udbredelsesformer og –retninger. Indirekte dokumenterede resultater (fx vandprøvetagning) af frakturingerings-støttet oprensning viser ofte umiddelbare oprensningsforbedringer efterfulgt af tilbageslag af høje forureningskoncentrationer. Dette indikerer at udførte sprækker på lang sigt ikke yder tilstrækkelig støtte til oprensningen. Fremadrettet bør de der implementer teknologierne til forbedret stoftilførsel, som en del af oprensningsdesignet, overveje hvilke sprække egenskaber, der er nødvendige for at opnå fastsatte oprensningsmål indenfor en rimelig tidshorisont, samt om lokale geologiske og geotekniske forhold understøtter disse.

Således er det nødvendigt, via den videnskabelige litteratur, at øge bevidstheden om lokale geologiske og geotekniske forholds indflydelse på dannede sprækkers egenskaber og dermed hvad der er opnåeligt med tilførselsmetoderne under forskellige forhold. Bidrag til direkte dokumentation af alle tre metoder på dybder overstigende 5 m u.t. er vigtige, da det på nuværende tidspunkt reelt set ikke vides om metoderne til forbedret tilførsel kan opfylde kravene for effektivt forbedret oprensning ved større dybder.

Der er derfor blevet udført et feltforsøg, der for første gang har haft til formål at udføre pilotforsøg og direkte dokumentation af alle tre metoder til forbedret tilførsel ved relevante dybder (2,5-9,5 m u.t.). Direkte dokumentation på større dybder var begrænset til kerneprøvetagning, men blev suppleret af udgravning ved lave dybder. En morænelerslokalitet i Danmark blev udvalgt til pilotforsøgene, da det er en almindeligt forekommende lavpermeabel, og ofte forurenet aflejring i Nord Europa og Nord Amerika.

Pilotforsøgene har vist at hydraulisk frakturering fungerer efter hensigten ved 3 m u.t. Forsøg på at udføre horisontale (tætliggende) sprækker ved 6-7 og 9,5 m u.t. lykkedes dog ikke. Sprækker udført ved pneumatisk frakturering (4-8 m u.t.) var umiddelbart horisontale, men tilbøjelige til afbøjning i naturlige (vertikale) sprækker i sedimentet. Tætte sprækkenetværk blev ikke observeret. Tynde, tætliggende sprækker kan dog muligvis opnås, hvis mindre afstand mellem fraktureringsdybder implementeres. Direkte sonde tilførsel formåede at skabe tætliggende, horisontale stoftilførsler ved alle afprøvede dybder (2,5-3,5, 6-7 og 8,5-9,5 m u.t.).

Resultaterne stemmer godt overens med forventningerne, givet de aktuelle geologiske og geotekniske forhold på den valgte lokalitet (normalkonsolideret, naturligt opsprækket basal till). Resultaterne efterviser ydermere nødvendigheden af grundig geologisk karakterisering og geotekniske forsøg ved alle *in situ* oprensningslokaliteter, som potentielt kunne støttes af forbedret tilførsel.

Uafklarede problemstillinger består. For pneumatisk og hydraulisk frakturering omhandler de 1) den nedre grænse for indbyrdes afstand mellem dannede sprækker ved større dybder end 5 m u.t. og 2) metodernes, især hydraulisk frakturerings evne til at skabe subhorisontale sprækker ved større dybder end 5 m u.t. For direkte sonde tilførsel er det uklart hvor stor betydning tilførselsvolumener har på tilførslernes orientering, form og radius.

List of contents

1 Introduction

Millions of contaminated sites exist all over the world (USEPA, 2004; EEA, 2007). In many cases, the sites pose a threat to the surrounding environment (air, surface water, ground water) and, hence, plant, animal and/or human life. Remediation is a necessity.

Remediation needs vary from site to site, as many different types of contaminants exist and have been spilled into many different types of natural environments. Spreading has occurred according to the properties of the contaminants and the given environment, resulting in highly variable contaminant distribution. Accordingly, many treatments and methods for their application have been developed.

The purpose of this PhD project has been to research possible enhancements of *in situ* mass removal of chloroethenes in clay till. This contaminant group and sediment type are both common (ATSDR, 2010; Sladen & Wrigley, 1983). Together, they represent a combination that is particularly difficult to remediate. Focus has been placed on the (enhanced) delivery of *in situ* remediation amendments in clay till, not the contaminants themselves, nor the specific *in situ* remediation amendments developed for their treatment. Numerous laboratory and field studies have proven the presently existing *in situ* mass removal treatments to be effective when sufficient contact between amendments and contaminants is provided (e.g. Cundy et al., 2008; Scheutz et al., 2008; Tsitonaki et al., 2010). In the words of Nyer and Page (2004), "the main problem we will face in remediation over the next decade is delivery". However, to understand the issues of distribution and consequent remediation challenges, the characteristics of the contaminants and of clay till must be recognized.

1.1 Chloroethenes and clay till

Chloroethenes are a commonly encountered group of contaminants due to their previously extensive use as solvents in dry-cleaning and metal industries in the 1960s and 70s (Kjeldsen & Christensen, 1996). They are now suspected carcinogens (vinyl chloride proven) with very low allowable concentrations in drinking water according to Danish regulations (0.2-1 ug/L, DEPA, 2009). Most chloroethenes are furthermore dense non-aqueous phase liquids (DNAPLs) with high viscosity and very low solubility in water (Parker et al., 1994). These characteristics allow them to migrate through sediments of high permeability as a separate liquid phase, leaving behind a trace of residual phase contamination (Figure 1.1a). On top of low-permeability layers (hydraulic conductivity $K < 10^{-6}$ m/s, Bures et al., 2004), the chloroethenes will form separate phase pools (Figure 1.1b). If left in place, the residual and pooled separate phases subsequently constitute long-term sources of contamination to infiltrating/passing groundwater due to their low solubility and high toxicity.

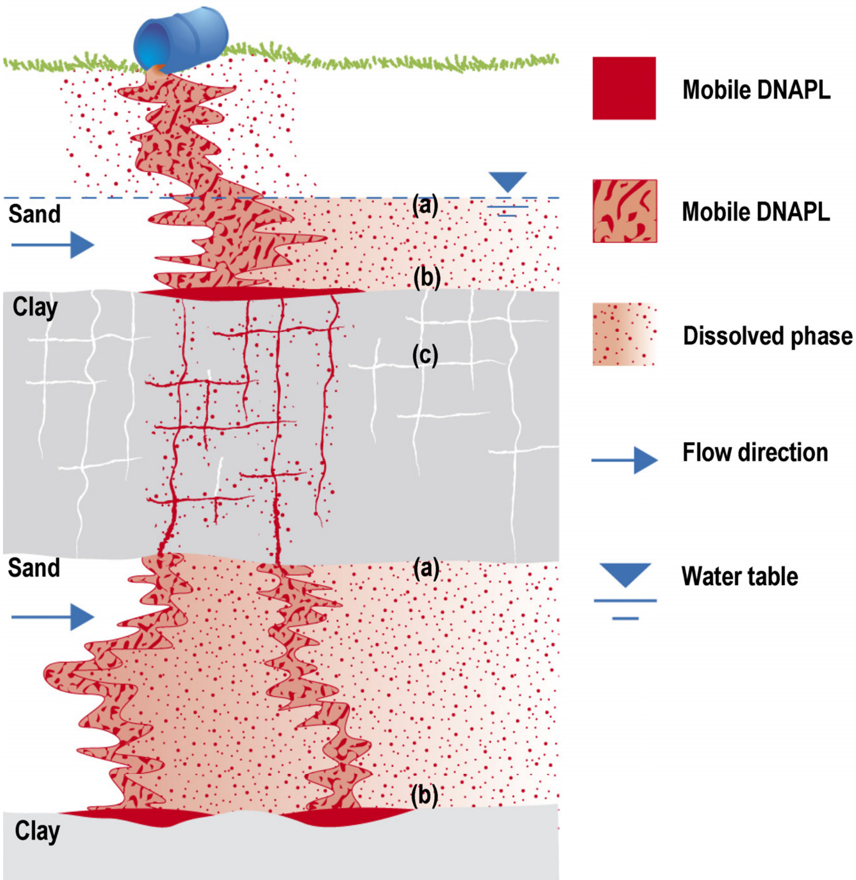


Figure 1.1: (a) DNAPL migration through a high-permeability sediment as a separate liquid phase, leaving behind a trace of residual phase contamination. (b) Separate phase pooling of DNAPL on top of a low-permeability layer (with hydraulic conductivity $K < 10^{-6}$ m/s). (c) Rapid lateral and vertical transport of DNAPL in natural fractures in a low-permeability clay till sediment. Figure adapted from Jørgensen et al. (2010).

Clay till is a commonly encountered low-permeability sediment, which is typically naturally fractured (Johnson et al., 1989; Parker et al., 1994; Parker et al., 1997; Klint, 2001; Jørgensen et al., 2004; Chapman & Parker, 2005). When a clay till is contaminated with chloroethenes, the fractures will serve as rapid transport conduits for the chloroethenes laterally within the sediment and to deeper layers (Figure 1.1c). However, part of the contaminant mass will also diffuse into the low-permeability matrix surrounding the fractures. Once separate phase DNAPL is no longer present in the fractures themselves (either due to migration or remediation), this residual mass will constitute a long-term source of contamination to infiltrating groundwater due to back-diffusion (Parker et al., 1994; Parker et al., 1997; Chapman & Parker, 2005), see Figure 1.2. In contrast to a residual mass in high-permeability sediments (and on top of low-permeability sediments), however, the back-diffusing mass lodged in low-permeability matrices constitutes a significant remedial challenge.

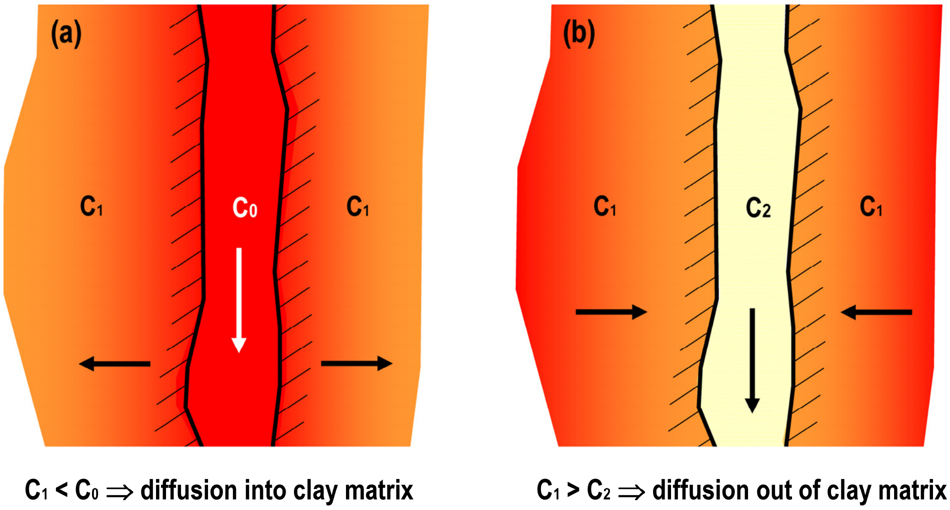


Figure 1.2: Principle of back-diffusion from a (DNAPL-)contaminated low-permeability clay matrix to natural high-permeability fractures (Parker et al., 1994; Parker et al., 1997; Chapman & Parker, 2005): (a) Early time - contaminant mass migrates downward through fractures and diffuses into the low-permeability matrix surrounding the fractures. (b) Late time - separate phase DNAPL is no longer present in the fractures themselves (either due to migration or remediation), and, hence, the residual mass in the low-permeability matrix diffuses out of the matrix back into the fractures.

1.2 Remediation options

Remediation applications are termed *ex situ* or *in situ*. *Ex situ* remediation involves excavation of contaminated soil and/or pumping of contaminated groundwater for treatment or disposal elsewhere (off-site). *In situ* remediation involves treatment of contaminants on site via mass transfer or mass removal methods. Mass transfer methods serve to extract contaminants from the subsurface (in gaseous or aqueous form) and subsequently treat them above-ground. Mass removal methods introduce amendments to treat contaminants in their actual location (in soil or water) in the subsurface.

Shallow soil contaminations that do not exceed 5 meters in depth below ground surface are mostly excavated and treated or disposed *ex situ* (e.g. Bures, 1998; USEPA, 2007). At greater depths, excavation of contaminant source zones is largely impracticable due to the costs (and security issues) associated with deep excavations. Deeper contaminations were previously widely remediated *ex situ* via pump and treat (P&T) of the resulting plumes in underlying groundwater aquifers (e.g. USEPA, 2007). Today, however, *in situ* remediation solutions are increasingly favored for a number of economic and/or practical reasons. For example, *in situ* remediation involves limited handling of contaminated soil (and water), minimization of site disruption and potentially adverse effects at ground surface, and, in some cases, fewer associated environmental impacts (USEPA, 2007; Lemming et al., 2010).

1.2.1 *In situ* remediation of chloroethenes in clay till

As it is indicated in the Section 1.1, severe problems with chloroethene contaminations in clay tills arise at greater depths than can practicably be excavated. P&T is also impracticable since chloroethene-plumes formed under a clay till source will be sustained for long periods of time due to back-diffusion (from the matrix to the fractures) and low solubility in groundwater (infiltrating via the fractures to aquifers below). *In situ* remediation is thus the only viable option.

Mass transfer technologies currently available for the *in situ* remediation of chloroethene contaminants include Soil Vapor Extraction (SVE; e.g. Hutzler et al., 1991), Thermal Desorption (ISTD; e.g. Heron et al., 2009), etc. Applicable *in situ* mass removal technologies include chemical reduction with zero-valent Iron (ZVI; e.g. Cundy et al., 2008), Chemical Oxidation (ISCO) with e.g. permanganate (Siegrist et al., 2001) or persulfate (Tsitsonaki et al., 2010), Enhanced Reductive Dechlorination (ERD; e.g. Sheutz et al., 2008), etc.

Goals of complete *in situ* mass removal are a necessity when the contaminants in question are chloroethenes. It has been shown that bulk residual mass removal may not effectuate a significant long term lowering of mass flux out of a source area because the mass remaining is sufficient to cause rebound of concentrations in percolating/receiving ground water (Chapman & Parker, 2005). However, just as P&T, *in situ* mass transfer methods are hampered by the diffusion-limited transport of chloroethenes to extraction wells placed in the source (or plume) area. For *in situ* mass removal methods, the diffusion-limitation on transport of contaminants and amendments makes it difficult to achieve sufficient contact between the two, also resulting in potentially impracticable remediation timeframes (e.g. Christiansen et al., 2008). Consequently, adequate *in situ* remediation of chloroethenes in clay till matrices will be provided by shortening the distances that the contaminants must travel to reach an extraction location or come into contact with amendments delivered *in situ*. Focus in this PhD project is placed on the challenge of delivering amendments at relevant depths and ensuring adequate distribution to eliminate persisting contaminant mass.

1.3 Natural and induced fractures

Distribution of *in situ* remediation amendments via the same natural fractures that have initially served to distribute contaminants will bring amendments into contact with the contaminant mass closest to the fractures. Once this mass has been remediated, however, the rate of further degradation will be dependent on the slow diffusion of contaminants out of the matrix to meet the amendments in the fractures (Chapman & Parker, 2005). Ideally, amendments will be able to reduce the distance that contaminants must travel by diffusing from the fractures into the matrix (as the contaminants initially did) to meet and degrade them there. This is a good start, but the typical frequency of natural fractures at depth (>1 m spacing; Klint, 2001; Chambon et al., 2009) is not high enough to adequately distribute *in situ* remediation amendments to ensure remediation within a reasonable time frame of 10 years. Modelling studies have suggested that spacing of fractures (containing ERD-amendments) must be 10 cm to achieve complete mass removal within this time frame (Chambon et al., 2010). I.e. much closer spacing of fractures/amendment distribution pathways is necessary.

Three technologies can be used for the purpose of enhancing delivery of *in situ* remediation amendments in low-permeability sediments: pneumatic fracturing, direct-push delivery, and hydraulic fracturing. Pneumatic and hydraulic fracturing have been used commercially for 15-20 years to enhance *in situ* mass transfer and mass removal methods (Christiansen et al., II). They are broadly accepted as viable and reliable methods for *in situ* remediation enhancement (USEPA, 1999; Roote, 2000; Schuring, 2002). However, tests of the hydraulic fracturing technology at Danish clay till sites have been largely unsuccessful (Walsted et al., 2002; Westergaard, 2005; Blem et al., 2006; Jørgensen et al., 2007; Christiansen et al., IV). A pilot test of pneumatic fracturing at a Danish clay till site similarly did not produce expected results (Christiansen et al., I). The capabilities of direct-push delivery are yet largely undocumented, but use of the method at several Danish clay till sites show promising results (Kjærsgaard, 2006a,b; Tsitonaki & Broholm, 2010; Christiansen et al., III).

1.4 Aim of the PhD project

The aim of the PhD project has been to shed light on the technologies that can enhance *in situ* mass removal of chloroethene-contaminated clay till sites: pneumatic fracturing, hydraulic fracturing, and direct-push delivery. Specific objectives have been to:

- Provide an overview of experiences to date with the technologies that can be used for enhanced delivery of *in situ* remediation amendments in clay till.
- Gain an overview of what these enhanced delivery methods can be expected to achieve based on geology and soil mechanics.
- Conduct field tests of the enhanced delivery methods at one site, and thus, uniquely, establish a basis for evaluation and comparison of the methods under similar conditions, using comparable procedures and documentation methods.

The objectives have been achieved via the activities listed in Table 1.1. A conceptual illustration of the conducted field tests, associated relevant contamination, and a shallow excavation (direct documentation) is shown in Figure 1.3.

1.5 Content of the PhD thesis

The contents of the individual sections of this PhD thesis are as follows. Section 2 describes clay till. Geological and geotechnical variations that are believed to be of importance in the perspective of enhanced delivery are discussed. Section 3 examines the technologies that can be used for enhanced delivery in clay till, what remediation goals they seek to meet, and parameters essential to remediation design. Furthermore, the important distinction between injection and fracturing, and soil mechanics theory necessary to understand fracture propagation patterns are presented. In Section 4, available documented results of the enhanced delivery methods are summarized, including the results of the pilot tests that have formed the backbone of the PhD project. Sections 2-4 are thus of a more general nature, emphasizing pilot test results obtained during the PhD project. A short ‘Findings’ subsection concludes each of these sections. Section 5, Discussion, focuses mainly on the comparability and representativity of these specific pilot test results along with the current status of the enhanced delivery methods. Section 6, Conclusions, naturally, concludes the thesis, while Section 7 offers suggestions for directions of further research within this area. The research results presented in the PhD thesis are a summary of four scientific papers, which are found in the appendices.

Table 1.1

Overview of PhD project aims, associated activities, duration, and participants

Aim	Activity	Year	Participants
Overview of experiences to date with the enhanced delivery methods pneumatic fracturing, hydraulic fracturing, and direct-push delivery	Literature review (incl. contact to researchers and vendors)	2005*-2009	Judith Wood Bertel Nilsson Knud Erik Klint William Slack Lawrence Murdoch Deborah Schnell Michael Liskowitz Mette Broholm
Overview of the expectable capabilities of the technologies as enhanced delivery methods based on geology and soil mechanics	Field course in geological characterization of clay till	2005*	Judith Wood Knud Erik Klint
	Course in soil mechanics	2007	-
	Literature study (incl. contact to researchers)	2005*-2009	Knud Erik Klint Niels Foged
Basis for evaluation and comparison of enhanced delivery methods via field tests	Pneumatic fracturing pilot test incl. direct and indirect documentation activities	2005*	Charlotte Riis Anders G. Christensen Judith Wood Stine Christensen Mette Broholm
	Supplementary direct documentation activities	2006	Charlotte Riis Anders G. Christensen Mette Broholm
	Direct-push delivery pilot test incl. direct documentation activities	2008-2009	Ida Damgaard Mette Broholm Kresten Andersen Palle Ejlskov
	Hydraulic fracturing pilot test incl. direct documentation activities	2008-2009	Ida Damgaard Mette Broholm Knud Erik Klint Bertel Nilsson Thomas Brøker
	Geological characterization	2005*-2010	Ida Damgaard Timo Kessler Knud Erik Klint Bertel Nilsson
	Cost survey	2009-2010	Michael Liskowitz Eliot Cooper Gordon Bures

*Activity commenced during MSc project conducted with Judith Wood.

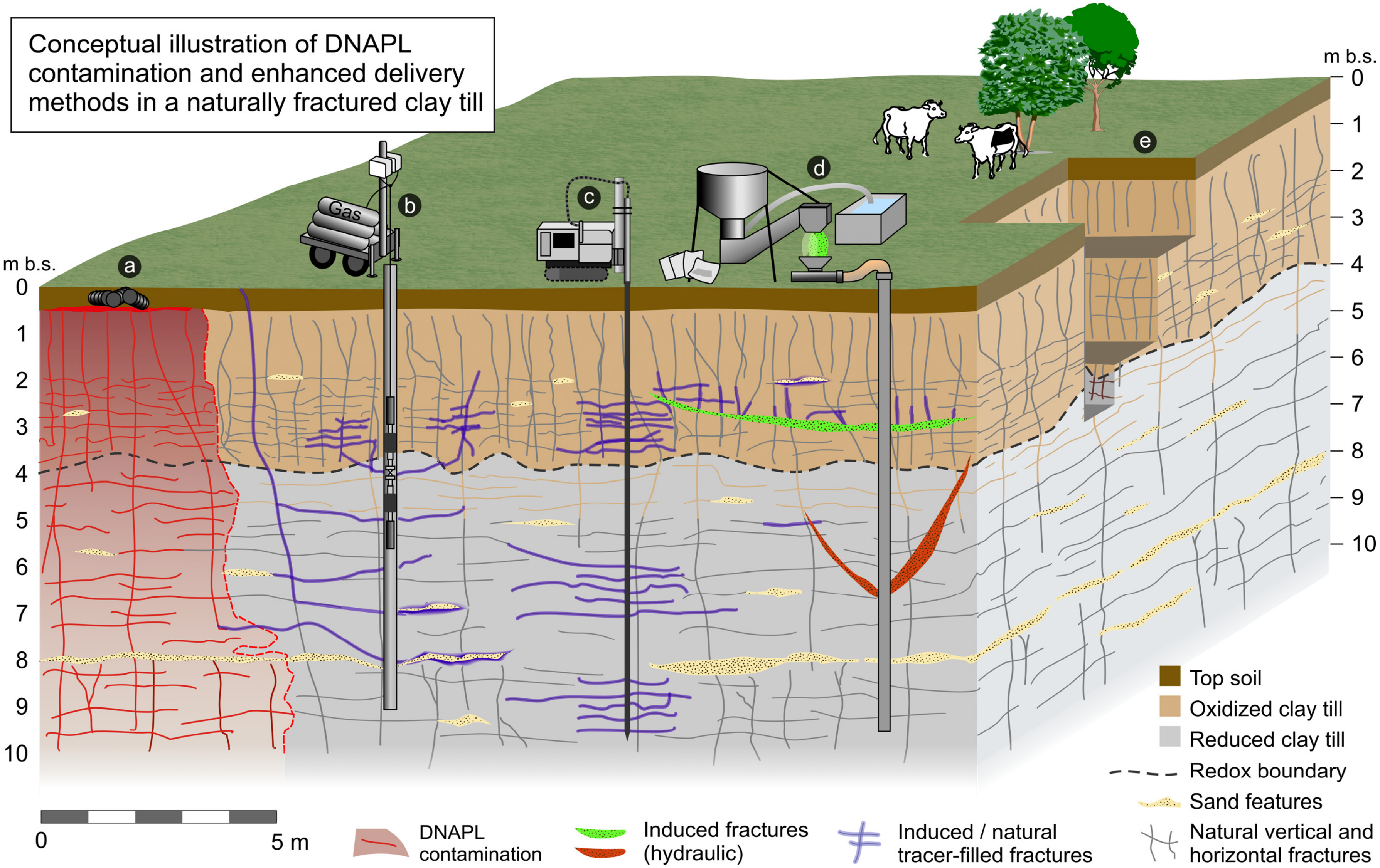


Figure 1.3: (a) DNAPL and residual mass contamination of a clay till. (b-c-d) Pneumatic fracturing, direct-push delivery, and hydraulic fracturing in a clay till as it was carried out and at the Vasby site. (e) Simple/shallow excavation of a clay till, illustrating its geological characteristics.

2 Clay till

Knowledge of the physical, chemical, geological and geotechnical/mechanical properties of contaminated deposits is valuable because it provides a fundamental understanding of their ‘typical’ characteristics. Based on these, initial estimates of advective flow and diffusion rates and thus potential remediation times may be obtained for contaminated sites that may be poorly characterized.

All the soil and rock types that may be encountered in the subsurface can be divided into six main categories: sedimentary deposits, organic deposits, carbonate deposits, evaporites, bedrock, and other deposits (Larsen et al., 1995). All can become contaminated, and those that have low permeability are particularly difficult to remediate. Focus here is placed on one type of low-permeability deposit, clay till, due to its predominance in North America and Europe (Sladen & Wrigley, 1983; Murdoch & Wilson, 1994; Klint & Gravesen, 1999), see Figure 2.1. Furthermore, clay till encompasses a geological and geotechnical variability that can exemplify the full spectrum of considerations necessary when assessing the suitability of a given contaminated site to undergo enhanced delivery of *in situ* remediation amendments.

Clay till is a sedimentary deposit, and while clay deposits may stem from a number of different geological time periods and depositional environments, the term ‘till’ is only used in reference to poorly sorted (diamict) deposits of glacial origin (Larsen et al., 1995). The clay content of clay tills ranges from a few

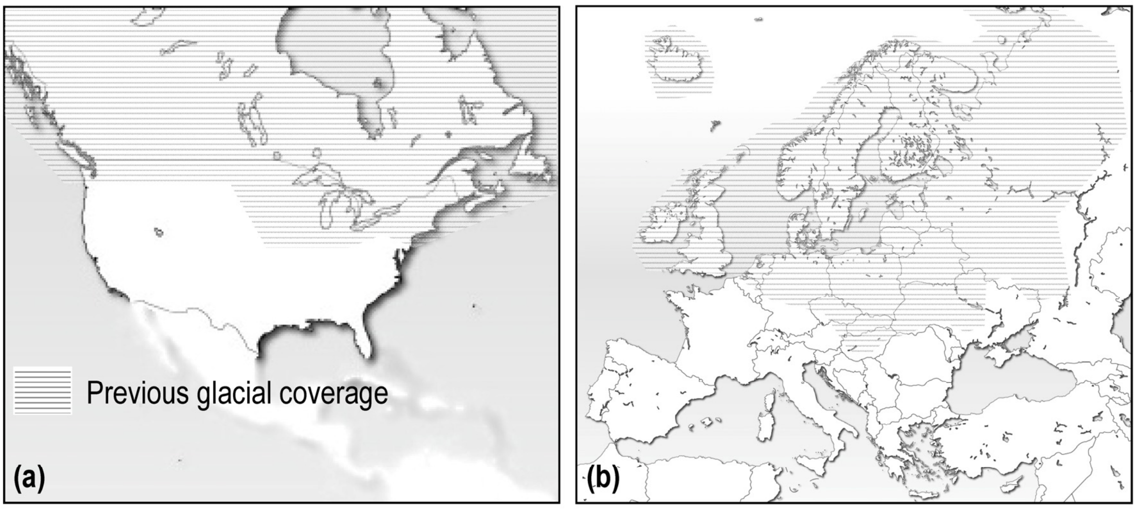


Figure 2.1: Pleistocene glacial coverage of (a) North America and (b) Europe. Adapted from Levin (2006). Previous glacial coverage implies glacial deposits, often clay till (Larsen et al., 1995).

percent to 35% (Houmark-Nielsen et al., 2005). Accordingly, reported porosity values range from 0.10-0.46, and hydraulic conductivity values range from 10^{-12} – $1.3.10^{-4}$ m/s (Cherry, 1989; D'Astous et al., 1989; Fetter, 1993; McKay et al., 1993a; Jørgensen et al., 1994; Jørgensen & Spliid, 1994; Parker et al., 1994; Sidle et al., 1998; Klint & Gravesen, 1999; McKay et al., 1999; Lindhardt et al., 2001; Nilsson et al., 2001; Walsted et al., 2002; Jørgensen et al., 2003; Iversen & Jakobsen, 2004; Styczen et al., 2004; Blem et al., 2006; Broholm et al., 2006; Riis et al., 2006a).

However, in terms of *in situ* remediation of contaminated clay till, knowledge of porosity and bulk hydraulic conductivity are not sufficient to evaluate potential remediation times. These deposits consist of a low-permeability matrix and high-permeability features, such as fractures. Transport in the low-permeability matrix is diffusion-limited (Johnson et al., 1989). The high-permeability features serve as the primary advective flow and transport conduits (Jørgensen et al., 2004). Hence, the spacing and apertures (Figure 2.2) of fractures are critical parameters in determining remediation timeframes. A number of Danish clay till sites have been assessed in terms of fracture presence, a summary is given in Table 2.1.

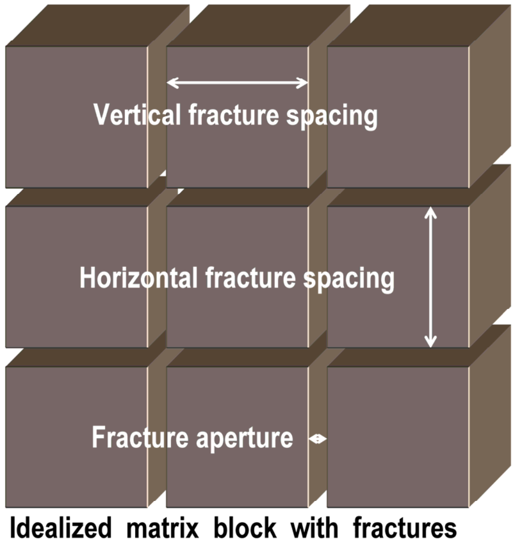


Figure 2.2: Definition of vertical and horizontal fracture spacing and fracture aperture in an idealized matrix block with fractures.

2.1 Clay till types

The large variations in fracture depths, apertures, and spacing illustrated in Table 2.1 are not unexpected in a deposit as heterogeneous, by definition, as clay till. However, different fracture types and frequencies are distinguishable in different types of clay till, which are classified as follows: basal (lodgement) till, flow till, melt-out till, and drop till (Klint, 2001). The classifications are based on the various glacial environments in which tills are deposited, as surface loading conditions are thought to have great influence on the types of till and hence formation of fractures and other features (folding, faulting, etc.).

Table 2.1
Summary of natural fracture observations in Danish clay tills

Parameter	Range of values	Average	# of observations	Values from 1 Canadian site[f]
Redox boundary [m b.s.][a]	2 - 6.5	4.2	24	4-6
Max. fracture depth [m b.s.][c]	2 - >9	> 5	19	10
Vertical fracture spacing at < ~5 m b.s. [cm][d]	0.5 - 667	83 (27)*	53 (45)*	2-100
Horizontal fracture spacing at < ~5 m b.s. [cm][d]	0.3 - 165	75 (15)*	52 (43)*	1-2
Vertical fracture spacing at > ~5 m b.s. [cm][d]	?	-	-	100-170
Horizontal fracture spacing at > ~5 m b.s. [cm][d]	?	-	-	60-170
Fracture aperture [μm][e]	31-3000	663	11	1-43

*The number stated in parentheses represents a more appropriate value/number, as a minor part of the observations with uncharacteristically large fracture spacings have been omitted.
[a](Houmark and Nielsen, 2005; Klint et al., 2001; Klint & Gravesen, 1999; Klint, 2004a; Klint, 2004b, Lindhardt et al., 2001, Blem et al., 2006)
[b](Klint, 2004a; Klint, 2004b)
[c](Klint, 2004a)
[d](McKay et al., 1999; Lindhardt et al., 2001; Christiansen et al., I; Nilsson et al., 2001; Klint & Fredericia, 1995; Sidle et al., 1998; Nygaard, 1999; Klint, 2004b; Jakobsen and Klint, 1999; Jørgensen & Spliid, 1998; Klint et al., 2001; Jørgensen et al., 2003; Klint & Gravesen, 1999)
[e](McKay et al., 1999; Jakobsen & Klint, 1999; Jørgensen & Spliid, 1994; Sidle et al., 1998; Nilsson et al., 2001; Christiansen et al., I)
[f](D'Astous et al., 1989, Cherry, 1989, McKay et al., 1993; Klint, 1996; Sidle et al., 1998)

Basal till is always deposited under a glacier, i.e. sub-glacially (American Geological Institute, 1984), and is the most common till type in North America and Europe (Sladen & Wrigley, 1983; Houmark-Nielsen et al., 2005). Flow and melt-out tills are typically associated with supraglacial, proglacial, and glacial margin environments, but subglacial melt-out tills can be found. Drop tills are formed only in pro-glacial environments.

Each till type can to some extent be linked to certain geotechnical and geological characteristics (Sladen & Wrigley, 1983; Foged & Steensfelt, 1992; Klint, 2001). In the perspective of enhanced delivery, some tills are suitable while others are decidedly unsuitable. In the following subsections, focus is placed on the geological and geotechnical features of clay till that are believed to have the most significant impact on enhanced delivery. These are natural fractures (and other high-permeability features) and consolidation. How and why their influence is asserted is subsequently discussed in Section 3.3 (Mechanics of fracturing).

2.2 Natural fractures in clay till

Natural fractures in clay till may be divided into 3 main groups: 1) glacial-tectonic fractures; 2) neotectonic fractures; and 3) contraction fractures (Klint, 2001; Klint et al., 2001, Klint, 2004a). Focus in the following is placed on subglacial-tectonic fractures and contraction fractures commonly observed in basal clay tills. The descriptions are exemplified with data from the Vasby clay till site, which was thoroughly geologically characterized by Christiansen et al. (I, III, and IV), see Figure 2.3. A summary is given in Table 2.2.

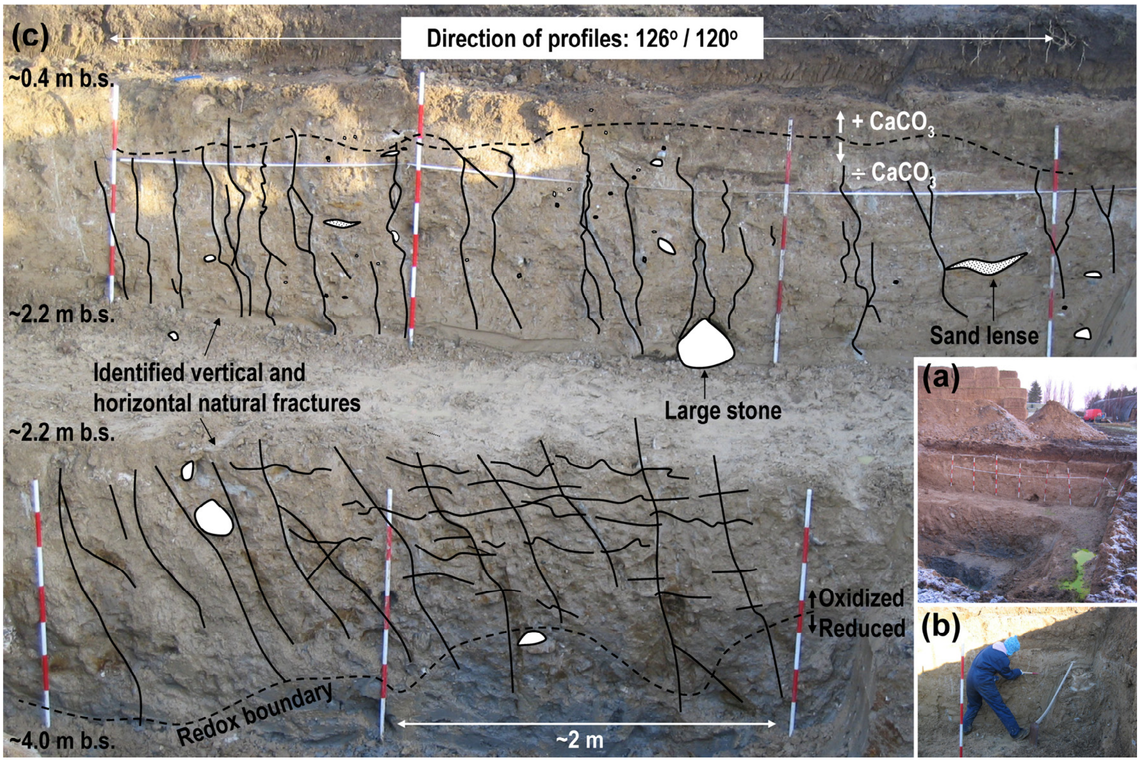


Figure 2.3: (a) One of the excavations conducted at the Vasby site. Area ~10x10 m, depth ~ 4.8 m. (b) Exposure of natural fractures via manual scraping. (c) Profiles of the excavation, where natural fractures, sand lenses, and redox boundary were identified. The average spacing of identified vertical fractures visibly decreases from the upper (~20 cm) to the lower profile (~30 cm). On the bottom profile (not shown, 4.0-4.8 m b.s.), spacing increased to ~2 m. The average spacing of identified horizontal fractures is ~10 cm.

Table 2.2

Orientation and characteristics of common natural fractures in basal clay tills (based on Klint et al., 2001; Klint, 2001; Klint, 2004a), exemplified via observations made in excavations at the Vasby site (Christiansen et al., I).

Fracture type		Orientation	Characteristics	Observations at the Vasby site
Subglacial-tectonic	*Subhorizontal shear*	- Sub-horisontal - Dip slightly (0-20°) toward or away from the direction of ice movement	- > 6 m long - Undulating surfaces w/ stripes - Sand- or silt-filled - Connect vertical fractures - Found in most lodgement tills - Present throughout till - Increased frequency toward bottom of till - Hydraulic properties poorly examined	- Observed from 2-4 m b.s. - Expected to continue throughout both till units (14-16 m total thickness) - Spacing: 20 cm
	Vertical shear	- Vertical / subvertical - 60-90° dip - Oriented perpendicularly to the direction of ice movement	- Sets of primary and secondary conjugated fractures (with acute intersection angle = 20°) - Planar form - Can traverse till deposits of thicknesses exceeding 10 m if underlain by well-drained deposits	- Observed from 0-4 m b.s. - Oriented NE-SW - Spacing (coupled w/ extension fractures) – 6-200 cm (increased from 2-4 m b.s.)
	Extension	- Vertical - 80-90° dip - Oriented parallel to direction of ice movement	- Are of primary importance for transport of various substances to groundwater	- Observed from 0-4 m b.s. - Oriented N-S - Spacing – see vertical shear
Contraction*	*Freeze-thaw*	- Subhorizontal - > 0.5 cm spacing	- Common/ever-present above water table - Decreased frequency over depth - Form zones with typical horizontal spacing of 0.5 cm (unsaturated in summer, saturated in winter – in this period basis for large lateral flow)	- Observed from 0-2 m b.s., likely present until 3.5- 4 m b.s. (coincident w/ redox boundary) - Spacing 0.87 cm at 1 m b.s. increased until cessation at redox boundary
	Desiccation	- Vertical - Irregular polygons	- Common/ever-present above water table - Decreased frequency over depth - Found only in fine-grained sediments (do not cut through larger sand layers/lenses	

* Found not only in basal clay tills. Freeze-thaw fractures are common in all cohesive (sedimentary) deposits, while desiccation fractures are common in all cohesive, fine-grained (sedimentary) deposits.

2.2.1 Subglacial-tectonic fractures

Subglacial-tectonic fractures arise from the glacier load and movement over a foundation of basal till. Thus, their orientation is systematically related to the directions of ice movement. Subglacial-tectonic fractures may be further divided into 4 groups: subhorizontal shear, conjugating (vertical) shear, (vertical) extension and hydro-fractures. Hydro-fractures are rare, and thus not discussed further. The spacing of the other three types of sub-glacial-tectonic fractures varies with depth and between deposits.

2.2.1.1 Vertical subglacial-tectonic fractures

The presence of more than two systems of vertical fractures at a site is attributable to more than one glacial advance or to neo-tectonic activity in the region (Klint et al., 2001; Klint, 2004a). A basal clay till is termed as type A (ductile) when it is unfractured, and as type B (brittle) when it is extensively systematically fractured.

Data from extensive coring at the Vasby site and existing borehole logs (for permanent well installations in the area) indicate that the site consists of two clay till units overlying a secondary sand aquifer, another clay till, and finally a primary limestone aquifer. The upper two till units have a combined thickness of 14-16 m and are separated by a discontinuous (melt water) sand layer at approximately 8 m b.s., varying in thickness from 0-1 m (Christiansen et al., IV). Excavations in the uppermost clay till at the Vasby site revealed two sets of systematic vertical fractures. Conjugating vertical shear fractures oriented NE-SW and vertical extension fractures oriented N-S were observed from 0-4 m b.s. with a horizontal spacing increasing with depth from 6-200 cm. The extent of systematic natural vertical fracturing indicates that the upper clay till is a basal clay till (type B; Christiansen et al., IV). The vertical fracture orientations indicate that this till was deposited during the Late Baltic Advance (Christiansen et al., I).

2.2.1.2 Influence of drainage conditions on vertical fracture formation

Field studies indicate that there is a connection between the type of sediment underlying a till and the depth and intensity of vertical glacial-tectonic fractures in the till (Chambon et al., 2009). The size and intensity of fractures in tills overlying permeable deposits (well-drained tills) are generally greater than those of fractures in tills overlying low-permeability deposits (poorly drained tills). When the till thickness becomes large, the drainage function of underlying permeable deposits becomes negligible (Klint et al., 2001; Klint, 2004a). This appears to be confirmed by Canadian field studies where the maximum depth of observed fractures does not generally exceed 10 m in thick (20 to 50 m thick) clay deposits in the Sarnia (Ontario) and the Montreal areas (Cherry, 1989). At the well-drained Duffins Creek site, also in Canada, conjugating shear fractures extending beyond 12 m below surface were observed (Klint, 2001). It must be noted that the previous presence of permafrost and/or supply of a large amount of melt-water to an underlying deposit that might otherwise be deemed of high permeability, could have lowered its permeability at the time of till deposition. Thus, determination of the deposits underlying a till is not a foolproof method for evaluating whether the overlying till can be expected to be fractured or not. But generally, a poorly drained till is also poorly fractured (Klint et al., 2001; Klint, 2004a).

Given the sand aquifer underlying the second basal clay till unit at the Vasby site, it is probable that systematic vertical fractures are well-developed in at least parts of this deposit (~8-16 m b.s.). It is not possible to verify natural fracture presence positively, as naturally occurring fractures are very difficult to observe in cores and borehole auger cuttings, which are the only data sources available at depths exceeding 5 m b.s. (Christiansen et al., I). Some of the cores collected at depths exceeding 8 m b.s. expanded greatly, however, indicating that the sediment is in places soft, or unconsolidated. The lower basal clay till at the Vasby site is therefore assessed as type A/B, i.e. partly fractured (Christiansen et al., IV).

2.2.1.3 Subhorizontal shear fractures

Subhorizontal shear fractures are most often created along with the till in the deforming bed. Therefore, these fractures are expected to be found throughout basal tills regardless of drainage conditions. Their spacing will be relatively constant throughout, even increasing toward the bottom, if the underlying deposit is sand (Klint, personal communication 2010). They may furthermore have significant unbroken trace lengths (> 6 m, Klint & Jakobsen, 1999).

At the Vasby site, sub-horizontal shear fractures were observed in the excavations from 2-4 m b.s. (4 m b.s. largest visible depth) with a vertical spacing of 20 cm. Based on the presence of sand underneath both of the major till units at the Vasby site, the subhorizontal shear fractures are expected to continue throughout both till units at the same constant vertical spacing (of 20 cm, Christiansen et al., I and IV).

2.2.2 Contraction fractures

Contraction fractures may be expected in all cohesive deposits, as they arise due to climatic change, which results in desiccation (i.e. dry-out) and/or freeze-thaw processes in the subsurface. The fractures are thus irregularly oriented vertical fractures or a dense network of small irregular fractures, respectively (Klint et al., 2001, Klint, 2004a). In the upper 2 to 3 m b.s., the fractures may be so frequent that the till texture becomes fissile (Klint, 2001). In practice, it may be difficult to distinguish between the formation processes, hence the general name, contraction fractures. The influence of climactic changes, and thus the presence of contraction fractures, is negligible beyond a certain depth. Typically, the number of contraction fractures decreases with depth. The maximum depth of penetration of contraction fractures usually coincides with the depth of the redox boundary, which is typically found at depths of 2-6 m b.s. in Danish till plains and deeper in elevated areas. The clear connection between contraction fracture penetration depth and the depth of the redox boundary gives rise to the rule-of-thumb that any fractures present under the redox boundary are glacial- or neo-tectonic in origin (Klint et al., 2001, Klint, 2004a).

At the Vasby site, contraction fractures were observed to a depth of 2 m, but likely persist until 3.5-4 m b.s. coinciding with the observed depth of the redox boundary. Their closest spacing was 0.87 cm at 1 m b.s. and increased until none were observable at 3.5-4 m b.s. (Christiansen et al., I).

2.2.3 Hydraulic activity of natural fractures

Only a small fraction (5-23%) of the natural fractures in clays are typically hydraulically active in their natural state (Klint, 2001; Klint et al., 2001; O'Hara et al., 2000; Jørgensen et al., 2003), yet these hydraulically active fractures (and other high-permeability features) constitute the main transport pathways for naturally percolating water and, potentially, contamination. As stated previously, *in situ* remediation efforts can rely on distribution of amendments in these same fractures, as a good correlation will generally be expected between the location of natural, hydraulically active fractures and the bulk mass of contamination in clay tills, because the contamination will have spread into the deposit through the hydraulically active fractures. However, a much shorter timeframe is typically afforded for the remediation a given contamination than the time it has had to distribute itself in the subsurface. Consequently, remediation efforts at naturally fractured, clay till sites may be aided substantially via additional fracturing opening/dilating previously inactive fractures and/or creating new fractures.

2.2.4 Other high-permeability features in clay tills

Presence of sand stringers or lenses is also a common characteristic of many clay tills. While these features are frequently observed in the field and may be noted in borelogs, they often become omitted from geologic profiles and general site descriptions. This is unfortunate, as the permeable stringers and lenses act as large fractures, where advective transport and reactions may take place. The statistic distribution of these high-permeability features is currently being researched (Kessler, personal communication 2010).

At the Vasby site, sand stringers and lenses with a limited spatial extent (<10 cm thickness and <2 m in length) were observed in the excavations. The stringers were highly permeable, but occurred too sparsely to build a connected network of preferential flow paths. Thus, vertical fractures are believed to control advective transport in the weathered horizon (0-5 m b.s.) (Christiansen et al., III).

2.3 Consolidation states of clay till

When sediments are initially deposited, the three principal stresses (σ_x, σ_y, and σ_z) are in equilibrium and equal to the overburden pressure. External forces such as glaciation, erosion, desiccation, excavation, etc. can change the stress fields (Suthersan, 1999). The *in situ* stress relationship in a given sediment can be expressed in terms of consolidation, given by the *in situ* stress factor K_0 (Kidd, 2001). When the horizontal and vertical stresses are balanced, the sediment is normally-consolidated ($K_0=\sigma_h/\sigma_v=1$). When the vertical stress exceeds the horizontal, the sediment is under-consolidated ($K_0<1$), and when the horizontal stresses exceed the vertical, the sediment is over-consolidated ($K_0>1$).

It is commonly stated that basal clay tills, due to the massive overburden pressure applied to them during their subglacial deposition, are overconsolidated (e.g. Blem et al., 2006). While this may often be true, it is not always the case (Boulton & Paul, 1976; Foged & Steensfelt, 1992). Strength and consolidation are dependent on a deposit's clay content, the effective stress placed on it during deposition, the extent of pore pressure dissipation during deposition, as well as void ratio decrease (induced via shear and creep) during and after deposition, and drying and wetting cycles after deposition (Boulton & Paul, 1976; Foged & Steensfelt, 1992; Edil & Mickelson, 1995; Bell, 2002; Christiansen et al., II).

The clay till at the Vasby site in Denmark was initially believed to be significantly overconsolidated, as it is a distinctive basal clay till exhibiting extensive evidence of shear (cf. the observed shear fractures) and underlain by sand (implying good drainage conditions). However, anisotropically consolidated, undrained triaxial tests conducted on intact core samples collected from depths of 3, 6 and 10 m b.s. in a geotechnical boring at the site led to the conclusion that K_0 values of approximately unity were valid for all three depths (Christiansen et al., IV).

Thus, it is not necessarily possible to couple geological and geotechnical characteristics, as geological classification is based on depositional processes, while geotechnical characteristics are based on depositional and post-depositional processes. I.e. the evaluation of suitability of a given sediment to undergo enhanced delivery must be based on all the processes that have influenced it (from its deposition until the present). The methods that allow the greatest level of certainty with regard to geologically classifying a given sediment and determining its consolidation are field-based geological characterization via excavation and triaxial tests, respectively. It should be noted that unconsolidated, undrained triaxial tests may significantly underestimate the undrained shear strength of (disturbed) soil samples. Anisotropically consolidated, undrained triaxial tests are more expensive, but provide more reliable data (Christensen et al., 1992).

2.4 Findings for clay till

- The Vasby clay till consists of two basal till units (with a combined thickness of14-16 m) separated by a thin, discontinuous sand layer at ~8 m b.s.
- Excavations at the Vasby site show that the upper basal clay till (type B) is extensively systematically fractured with three vertical fracture systems (with an expected spacing of ~2 m) and many subhorizontal shear fractures (with a spacing of ~20 cm) traversing the till.
- Excavations at the Vasby site also show that sand lenses and stringers are present in the upper till but occur too sparsely to build a connected network of preferential flow paths. Thus, vertical fractures are believed to control advective transport in the weathered horizon (0-5 m b.s.) of the upper Vasby till.
- Cores collected at the Vasby site indicate that the lower basal clay till (type A/B) is soft (expanding) in places and firm in others, and thus likely systematically fractured to some extent. Spacing of potential natural fractures has not been determined.
- Triaxial tests indicate that the Vasby basal clay tills are normally consolidated (with K_0 ~1).

3 Methods for enhanced delivery in low-permeability media

Various technologies can be used to enhance delivery of substances in low-permeability media. In clay till and other clay-type sediments the options are hydraulic fracturing, pneumatic fracturing, and direct-push delivery (Murdoch & Wilson, 1994; Schuring, 2002; Christiansen et al., III), see Figure 3.1.

Fracturing was initially developed as a technology for enhancement of mass extraction in the oil industry (Murdoch & Wilson, 1994). Here, (vertical) fractures are emplaced in reservoirs at depths of several hundreds of meters to kilometres. The technology is termed environmental fracturing in an adapted form used for soil and groundwater remediation at comparatively shallow depths (Christiansen et al., II). With both environmental fracturing methods, a fluid is introduced to the subsurface under relatively high pressures (5-10 bar) to create a fracture (Murdoch & Wilson, 1994). Hydraulic fracturing utilizes a water-based slurry of guar gum gel and sand for emplacement of distinct, thick (~1-2 cm), sand-filled fractures (Figure 3.1b), while pneumatic fracturing utilizes a gas (typically nitrogen or air) to create allegedly bifurcating, thin, unpropped fractures (Figure 3.1c; USDoE, 1998).

Previously, the use of environmental fracturing was most commonly coupled with *in situ* mass transfer technologies in low-permeability settings. However, due to the development of seemingly effective *in situ* mass removal technologies (e.g. Zhang et al., 2003; Scheutz et al., 2008; Tsitonaki et al., 2010) and their distinct advantage of causing less site disruption, environmental fracturing is increasingly utilized to enhance delivery of *in situ* remediation amendments in low-permeability deposits (Bures et al., 2004). However, as the fracturing technologies can be utilized for mass transfer enhancements also, they should not be perceived as enhanced delivery methods only.

Direct-push delivery, on the other hand, is strictly an enhanced delivery technology. It utilizes direct-push apparatus (typically a GeoProbe®) to drive a probe into the subsurface. Upon reaching the desired depth, the probe can then directly deliver substances, e.g. *in situ* remediation amendments (Figure 3.1d). The direct delivery is a significant point of distinction from the fracturing methods that require the use of a borehole to obtain access to the desired fracturing depth.

With hydraulic fracturing, delivery of amendments can occur in three ways, depending on the properties of the amendment: 1) aqueous amendments can be injected into fractures subsequent to fracture emplacement (e.g. Christiansen et al., IV), 2) aqueous and solid amendments can be mixed into and introduced simultaneously with the hydraulic fracturing sand-slurry (e.g. Martin et al., 2002), or 3) solid amendments can entirely replace the fracturing sand so the resulting fracture is propped by amendments alone (e.g. Siegrist et al., 1999). With pneumatic fracturing, substances are delivered simultaneously with or immediately after fracture propagation (on atomized form). Neither pneumatic fracturing (ARS, 2010), nor direct-push delivery are suited for delivery of larger solid amendments, but can deliver nano- to microscale solids.

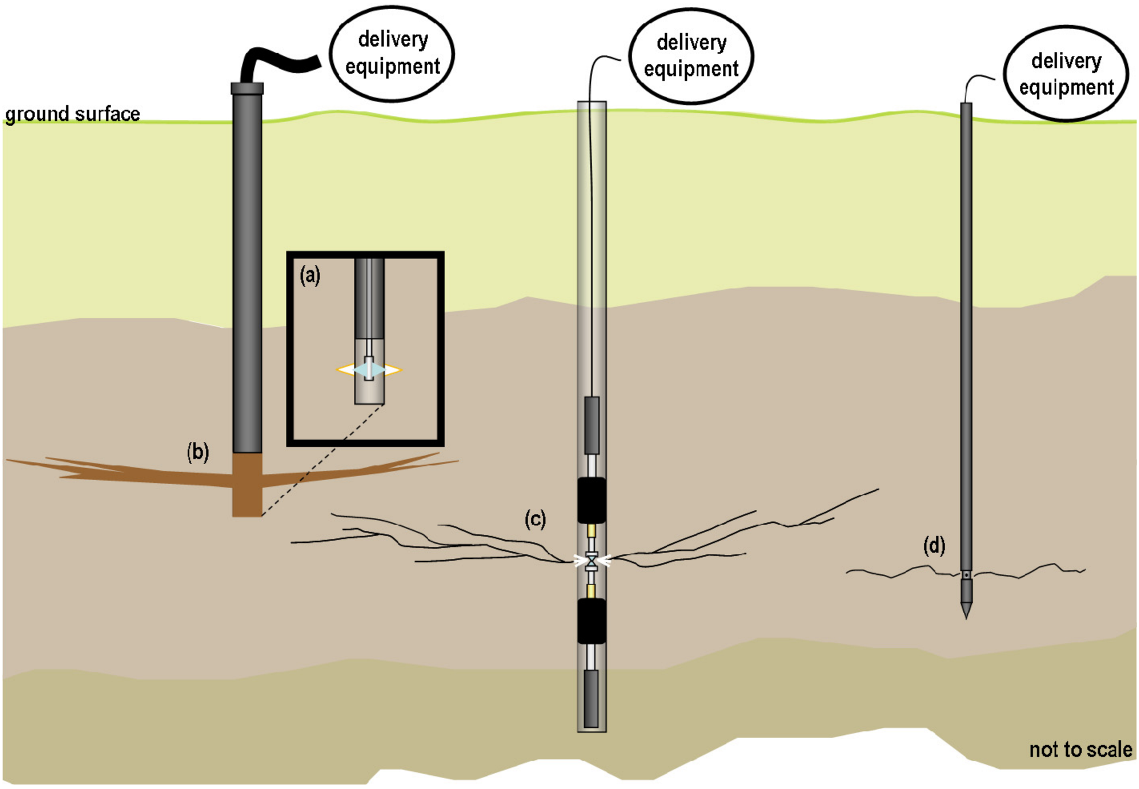


Figure 3.1: The enhanced delivery methods available in clay till, modified from Christiansen et al. (II). (a) Hydraulic fracturing proceeds by drilling a cased borehole, retracting the casing a little, and cutting a notch at the desired fracturing depth. (b) Subsequently, a sand-guargel-slurry is pumped into the borehole to initiate and propagate a large-aperture, sand-propped fracture (USEPA, 1995). (c) Pneumatic fracturing proceeds by drilling a borehole, retracting or removing the casing, and inserting a packer system to seal off the desired fracturing depth. A gas (nitrogen or air) is then introduced to initiate and propagate a small-aperture, unpropped fracture (USDoE, 1998). (d) Direct-push delivery proceeds by inserting a delivery probe into the subsurface and driving it to the desired delivery depth. Upon reaching the desired depth, substances are delivered directly (Christiansen et al., III).

3.1 Delivery via fracturing vs. injection

It is important to note that the method referred to here as direct-push delivery is often termed direct-push injection (e.g. Cooper et al., 2008). Actual injection only occurs if the sediment receiving a substance (via direct-push delivery) has a high permeability. In low-permeability sediments, injection is extremely slow if not impracticable, and attempts to increase injection rates typically require increased injection pressures. Specific levels depend on the given sediment.

The increased pressures lead to fracturing of the sediment, and, hence, substances are distributed in newly induced fractures (Bures, 2009). The findings of Christiansen et al. (III, IV) support the claim that direct-push delivery distributes substances via fracture inducement in low-permeability settings. Nonetheless, the method will here be referred to as direct-push delivery, not fracturing, as it is an injection method when utilized in high-permeability sediments, and the term delivery may encompass both.

The practical distinction between the enhanced delivery methods lies in the distribution of the injected fluids in the subsurface. Induced fracture orientations, forms, distribution radii, apertures (and/or created reaction zones), and spacing are crucial parameters to remediation design, estimation of remediation time frames, and, hence, comparison of the three methods' suitability at given sites.

3.2 Aims of enhanced delivery

In situ remediation is not typically an economically viable option at depths shallower than 5 m b.s., as soil to this depth can easily be excavated (and treated *ex situ*; Bures, 1998). Typically, *in situ* remediation is used at sites where the bulk of contaminant mass is situated at depths exceeding 5 m b.s. It is therefore imperative that fractures can be emplaced/induced here.

In the context of *in situ* remediation, the creation of horizontal fractures for delivery of amendments is desired. Vertical fractures (i.e. fractures inclining steeply upwards from their initiation point) may mobilize contaminants, with a risk of downward movement (Murdoch & Wilson, 1994). The formation of horizontal fractures facilitates the use of enhanced delivery methods to create permeable reactive barriers underneath contaminated soil volumes and, hence, a reduction of the vertical contaminant mass flux. The creation of closely-spaced horizontal fractures facilitates effective distribution of *in situ* remediation amendments within contaminated soil volumes (e.g. source zones). As mentioned in Section 1.3, 10-cm-spacing of fractures filled with ERD-amendments (assumed to be non-diffusive) is necessary to achieve complete contaminant mass removal in a clay matrix saturated with PCE[1] within a time frame of 10 years (Chambon et al., 2010).

3.2.1 Dependency on *in situ* remediation amendment properties

The goal of achieving a fracture spacing of 10 cm is a conservative requirement for remediation success. Fracture spacing requirements may be less strict, depending on the *in situ* treatment selected at a given site. This is due to the fact that some *in situ* remediation amendments are able to diffuse into the sediment, creating significantly wider reaction zones than the induced fractures themselves.

ZVI does not diffuse into the sediment matrix. When emplaced via hydraulic fracturing discrete reactive fractures were created (Siegrist et al., 1999). Permanganate, on the other hand, has been shown to diffuse up to 1.4 cm into a clay matrix within 20 days in laboratory experiments (Hønning et al., 2007) and up to 20 cm within 10 months under field conditions (Siegrist et al., 1999). Tsitonaki et al. (2010) state that heat activation facilitates diffusion of persulfate into a contaminated zone, but quantitative figures have not been obtained. With ERD, utilized substrates (e.g. molasses or soybean oil; Damgaard et al., 2009) will diffuse into low-permeability media. It is yet unknown, however, whether the dechlorinating bacterial cultures can diffuse into a low-permeability clay matrix. Results from Jørgensen et al. (2007) indicate this, but it is suggested that the bacteria may be transported in micro-fractures extending from emplaced hydraulic fractures. Similar results were obtained by Christiansen et al. (2008; Figure 3.2) with concentration profiles of chloroethenes suggesting a reaction zone width of ~10 cm around two natural fractures into which *Dehalococcoides* and substrate (emulsified soybean oil) were delivered approximately two years earlier.

3.2.1.1 Amendment-comparable tracers

Pilot tests to investigate the efficiency of a given *in situ* mass removal remediation strategy assisted by enhanced delivery can be conducted with tracers that have similar characteristics and behavior to amendments, rather than actual amendments. The advantages are reduced costs and increased visibility of substance distribution in direct documentation efforts, etc. So-called amendment-comparable tracers that are suitable under field conditions are rhodamine WT and fluorescein (Christiansen et al., I, III, and IV). Both are fluorescent and clearly

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[1] Tetrachloroethylene, the mother compound of many chlorinated solvent contaminations.

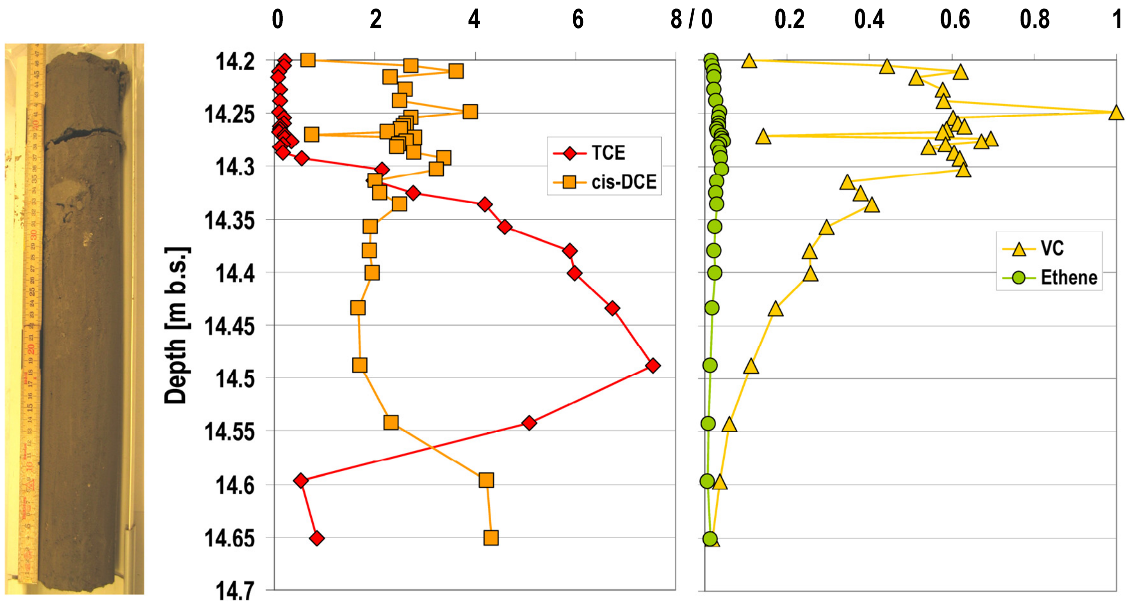

Figure 3.2: Concentration profiles for chloroethenes TCE (mother product), DCE, VC, and ethane in a core from a naturally fractured clay till treated via ERD. Modified from Christiansen et al. (2008). Two small-aperture natural fractures are present at 14.23 and 14.28 m b.s. ERD-amendments (D*ehalococcoides* bacteria and substrate) were delivered into these fractures two years prior to the core-sampling. The result was a degradation zone of ~10 cm, where TCE concentrations are significantly reduced, mainly due to degradation (sequentially via DCE) to VC.

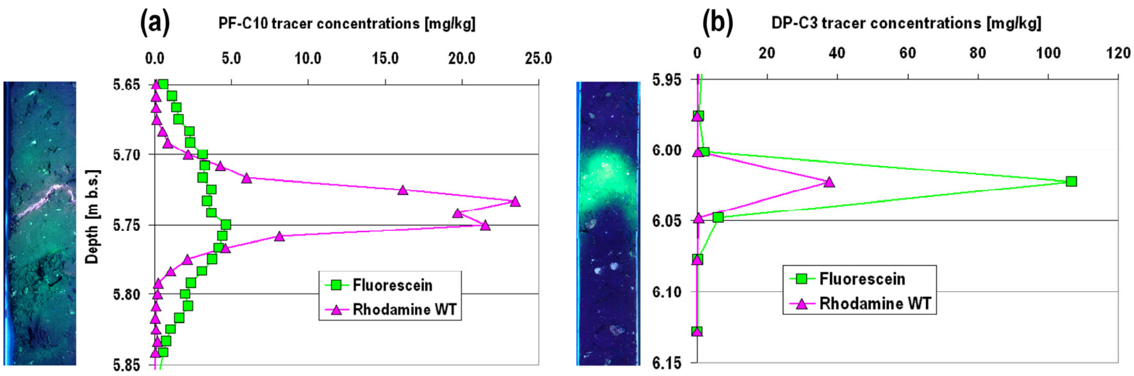

Figure 3.3: (a) Fractures induced during pneumatic fracturing at ~4.3 m b.s. in a Danish clay till. Tracers rhodamine WT and fluorescein were delivered during fracturing (resulting concentrations of both was ~10 000 mg/L) and make the induced fractures clearly visible in cores (and excavation) under 312 nm UV-light. (a) Fracture induced during direct-push delivery at ~6 m b.s. in the same Danish clay till. Tracers rhodamine WT and fluorescein were delivered (at concentrations of 2 000 and 10 000 mg/L, respectively). Only fluorescein is clearly visible due to the reduced concentration of rhodamine WT. Figure adapted from Christiansen et al. (I and III).

visible under 312 nm UV-light. The first sorbs strongly to the surfaces of fractures induced during enhanced delivery (Sabatini & Al Austin, 1991), mimicking non-diffusive *in situ* remediation amendments (ZVI). The latter is highly mobile and diffuses readily (Sabatini & Al Austin, 1991) into the low-permeability clay matrix from induced fractures, mimicking diffusive *in situ* remediation amendments (e.g. permanganate). See Figure 3.3. Whether (dechlorinating) bacteria belong to the first or second group must be researched further. Laboratory tests indicate that the tracers severely inhibit *Dehalococcoides ethenogens* bacteria (Riis et al., 2006b). Tracer pilot tests should therefore not be conducted within an area targeted for ERD-remediation.

3.3 Mechanics of fracturing

The following discussion is relevant for all three enhanced delivery methods. However, literature discussing the mechanics of direct-push delivery have not been located. The text is therefore based on hydraulic and pneumatic fracturing literature.

3.3.1 Influence of consolidation on fracture orientation

While the mechanics behind soil fracturing are disputed (Alfaro & Wong, 2001; Zhang et al., 2008), but there is general agreement that the pressure required to initiate a fracture must exceed the cohesive (tensile) strength of the formation and the overburden pressure (a function of the density and depth). Once initiated, fractures tend to form in the direction normal to the least stress (Murdoch & Wilson, 1994; Suthersan, 1999; Figure 3.4a). As a horizontal distribution of amendments is desired, overconsolidated sediments (where the principal stress is least in the vertical direction, i.e. $K_0 = \sigma_h/\sigma_v > 1$) are good candidates for

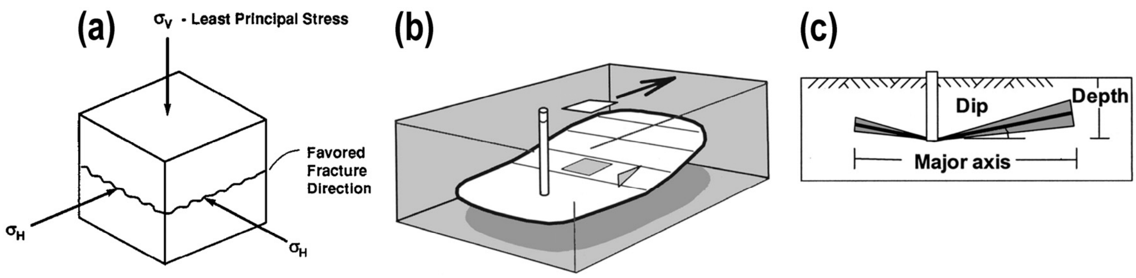

Figure 3.4: (a) Preferred (initial) horizontal fracture propagation direction upon fracturing in a sediment where the least principal stress is vertical (Suthersan, 1999). (b) Idealized form of a hydraulic fracture in an overconsolidated sediment (Murdoch & Slack, 2002). (c) Orientations and asymmetry of idealized hydraulic fracture (Murdock & Slack, 2002).

enhanced delivery of *in situ* remediation amendments, as horizontal fractures are expected to form.

The idealized form of a hydraulic fracture in overconsolidated sediments is well-documented at shallow depths (0-5 m b.s.) It constitutes an elliptical shape that is off-center from the fracturing borehole, representing a preferred direction of propagation (Murdoch et al., 2002; Figure 3.4b). It is sub-horizontally oriented, rather than completely flat-lying, as the idealized form curves towards the surface seeking pressure relief, taking on the shape of a shallow- to steep-sided bowl extending outwards from the initiation point (Murdoch & Wilson, 1994; Murdoch, 1995; Schuring, 2002; Murdoch & Slack, 2002; Figure 3.4c). Pneumatic fractures would be expected to exhibit similar general characteristics, but have not been documented systematically. However, rather than large, distinct fractures, a network of smaller-aperture fractures via extension of existing fractures and creation of secondary networks of fissures and channels is alleged (Markesic, 2000; USEPA, 1995; USDoE, 1998; Strong et al., 2004). Direct-push delivery also lacks systematic documentation (Cooper et al., 2008).

Despite the expectation of consolidation control over initial fracture propagation directions, few studies have coupled geotechnical measurements with documentation of induced fractures. The limited data available are summarized in Table 3.1, and suggest that factors other than consolidation influence fracture propagation.

3.3.2 Leak-off and short-circuiting

The possible lateral extent (or radius) of induced fractures is controlled by the injected volume and rate of leak-off, as fluid injected into the subsurface to initiate and propagate a fracture leaks out of the fracture (Murdoch and Wilson, 1994) at a rate that depends upon the permeability of the formation, the fluid rheology and the fluid pressure in the fracture. The effect of leak-off is one of the major differences between hydraulic and pneumatic fractures, largely because of the differences in viscosity of the injected fluids (aqueous and gaseous respectively). Localized high-permeability zones will cause localized leak-off. Thus, if a fracture encounters a sand lense or borehole, the leak-off that occurs in the lense may preclude further propagation.

An ideally formed fracture with a certain radius is the expectation when delivering a known amount of fluid with either fracturing method to an overconsolidated deposit. However, as noted in Section 3.3.1, it is important to recognize that other factors affect the orientation and radius of

Table 3.1
Summary of fracturing studies involving geotechnical tests

Study and sediment type	**Measured geotechnical parameters**	**Observed induced fracture orientation**
Laboratory study with artificially prepared sand-kaolin clay mix (Alfaro & Wong, 2001)	K_0 = 0.8 (OCR = 3) K0 = 1.2 (OCR=6) K0 = 1.4 (OCR=8)	Vertical Subhorizontal Horizontal
Hydraulic fracturing pilot test in silt/clay till (Wong & Alfaro, 2001)	Attempts to secure undisturbed soil samples for geotechnical testing failed	Fractures induced at 3, 3.5, 4, and 4.5 m b.s. and documented via excavation. Subhorizontal propagation confirmed.
Hydraulic fracturing pilot test in clay till, Næstved (Walsted et al., 2002)	OCR=6 and 14 at 2.5 m b.s. OCR=10 at 3.5 m b.s. OCR=6 and 7 at 4.5 m b.s.	Two fractures induced at 4 and 4.2 m b.s. were documented via coring. One propagated subhorizontally to one side, and inclined significantly upward (45°) to the other side. The other fracture initially propagated downward, and then steeply upward (55°).
Hydraulic fracturing pilot test in clay till, Haslev (Blem et al., 2006)	OCR = 2 at 2.4 m b.s. OCR = 4 at 4.3 m b.s. OCR = 4 at 6.3 m b.s. OCR = 3 at 8.2 m b.s.	Two fractures were induced at 4.5 and 8 m b.s. and documented via coring. Both propagated downward, and lack of fracture findings in most cores indicate erratic form.
Pneumatic fracturing pilot test in clay till, Vasby (Christiansen et al., I and IV)	K_0 = 1.10 at 3 m b.s. K_0 = 1.19 at 6 m b.s. K_0 = 0.83 at 10 m b.s.	Fracturing at 4, 5, 6, 7, and 8 m b.s. documented via excavation and coring. Indications are that fractures propagated subhorizontally initially but were prone to short-circuiting in natural vertical fractures.
Direct-push delivery pilot test in clay till, Vasby (Christiansen et al., III and IV)	K_0 = 1.10 at 3 m b.s. K_0 = 1.19 at 6 m b.s. K_0 = 0.83 at 10 m b.s.	Delivery at 2.5-3.5, 6-7, and 8.5-9.5 m b.s. documented via excavation and coring. Subhorizontal propagation. Some distribution in natural vertical fractures, seemingly without reduction in delivery radius.
Hydraulic fracturing pilot test in clay till, Vasby (Christiansen et al., IV)	K_0 = 1.10 at 3 m b.s.	Fracture induced at 3 m b.s. documented via excavation: subhorizontal with ideal form.
	K_0 = 1.19 at 6 m b.s.	Fractures induced at 6-7 m b.s. documented via cores. Few fracture findings indicate moderate to steep inclinations and erratic forms.
	K_0 = 0.83 at 10 m b.s.	Fracture induced at 9.5 m b.s. vented at surface, i.e. propagated vertically. Confirmed by shallow excavation.

induced fractures (particularly where K_0 ~1 / OCR ~5). Overconsolidation only suggests an *initial* fracture propagation of predominantly horizontal direction. When propagation is not altogether stopped by fracture intersection with high permeability zones or geological weaknesses, it is likely to be diverted from its original path (Murdoch & Wilson, 1994; Schuring, 2002). This phenomenon is sometimes termed short-circuiting (e.g. Markesic, 2000). Fractures can short-circuit along e.g. natural (vertical) fractures (Christiansen et al., I), bedding planes (Murdoch & Slack, 2002), and man-made features such as wells, utility lines, building foundations, etc. (Nilsson et al., 2000). Large vehicles also induce stresses that may affect fracture pathways (Murdoch & Wilson, 1994).

3.3.3 Preliminary guideline for fracture propagation patterns

Thus, the orientation of induced fractures can only crudely be estimated based on the consolidation of a given sediment. Without extensive knowledge of geological characteristics of that sediment, these estimates should be used with caution. Gathering of comparable geological and geotechnical data sets has not been the norm at fracturing sites since environmental fracturing emerged 15-20 years ago, but based on the limited data that are available, Christiansen et al. (II) propose a tentative guideline regarding fracture propagation patterns in clay-type sediments in Box 3.1.

To use the guideline, consolidation conditions and significant geological characteristics must be known at relevant depths for a proposed fracturing site. While it is believed that fractures induced pneumatically, hydraulically and via direct-push delivery, will largely conform to the guideline, recent field data from clay till suggest that the orientation of hydraulic fractures in normally to slightly overconsolidated clay till are highly erratic at depths exceeding 5 m.b.s. (Christiansen et al., IV).

3.3.4 Inducing new fractures vs. opening existing, natural fractures

It has been established that new fractures may not always form as a result of the fracturing process, or may have a very small horizontal radius, as propagation paths are often diverted along bedding planes, sand features, natural fractures, etc. It is important to consider the consequences that this may have on the ability of fracturing to overcome diffusion limitations on mass removal during *in situ* remediation efforts. Particularly the distinction between creation of new fractures, opening/dilation of existing fractures to hydraulic flow, or a combination of the two, is significant.

The degree of lasting remediation enhancements achievable via fracturing is extensive if facilitated by inducement of new fractures. Supplementary activation of (many) previously inactive fractures in the volume of contaminated soil is also beneficial. Both new and newly opened fractures can serve to improve access to the volume of contaminated soil by significantly reducing the length of diffusion pathways, and hence the diffusion limitations on mass transfer/removal. However, if fracturing facilitates distribution of amendments in natural fractures only, then the ability of fracturing to overcome diffusion limitations in a given sediment will depend entirely upon the frequency of natural fractures in the relevant remediation depth interval. As it has been described in Section 2.2, natural fractures are typically abundant in clay tills above the redox boundary (in the weathered horizon), whereas fracture frequency below this boundary (unweathered horizon) is highly variable.

Box 3.1
Tentative guideline for fracture propagation patterns in clay-type sediments

- *In under-consolidated sediments ($K_0<1$)*, geotechnical considerations imply that induced fractures will be steeply inclined, although well-developed bedding or lamination may cause the dip to be shallow.

- *In normally-consolidated sediments ($K_0 \sim 1$)*, the inclination of fractures will be controlled largely by geologic structure, such as bedding or pre-existing fractures. Other factors such as the fluid density, pressure gradients along the fracture, and interaction with the ground surface could also be important under these conditions.

- *In overconsolidated sediments ($K_0>1$)*, induced fractures are expected to be sub-horizontal, and mechanical interaction with the ground surface will likely cause upward propagation into a bowl-shape. Geological features that are mechanically weak (sometimes termed as "paths of least resistance"), e.g. natural horizontal and/or vertical fractures may, however, still affect propagation paths.

3.4 Findings for enhanced delivery

- In low-permeability media, some form of enhanced delivery is necessary if complete *in situ* remediation of residual chloroethene mass within a practicable timeframe is desired.
- Fracturing is inevitable during enhanced delivery of *in situ* remediation amendments in low-permeability media due to the pressures required to distribute amendments here.
- Typically, relevant depths for *in situ* remediation of chloroethenes in clay till will be at depths greater than 5 m b.s. To provide effective remediation, delivery of *in situ* remediation amendments must occur in horizontal fractures with close vertical spacing (~10-25 cm depending on chosen remediation methods).
- Consolidation plays an important role in induced fracture orientation, but many other (geological) factors are also influential. A tentative guideline for expected fracture propagation patterns in clay-type settings is proposed (Box 3.1).

4 Documented results of enhanced delivery methods

A thorough review of the literature discussing environmental fracturing in clay-type settings has been conducted (Christiansen et al., II). The review reveals that while more than 80 clay-type environmental fracturing sites are mentioned in summary reports and conference abstracts (e.g. Roote, 2000), the descriptions of most are very sparse. At least 30 fracturing applications have involved fracturing at 5-25 m b.s. It is difficult to state a precise number, as some applications are described so sparsely that not even fracturing depths are given. Empirically-based observations have been given preference in this review, as many published modeling studies on the propagation of fractures are focused on applications in deep hydrocarbon reservoirs. While the mechanics presented in these studies are relevant, the fractures simulated are predominantly vertical. Furthermore, the mechanism behind soil fracturing is disputed in hydrocarbon and environmental fracturing modeling studies alike (e.g. see review of fracturing mechanisms in soils by Alfaro & Wong, 2001, and Zhang et al., 2008). Directly documented characteristics of induced fractures are summarized in Table 4.1, and will be discussed briefly in the following subsections.

Documentation of delivery success and guidelines for design of delivery schemes have yet to be developed for direct-push delivery (Cooper et al. 2008). Results from a small number of applications in Danish clay tills, currently only published in Danish consultant reports, have therefore been included.

Only one field study encompassing pilot tests of all three enhanced delivery methods at the same site has ever been conducted (Christiansen et al., II and IV), see Figure 4.1. The tests focused on direct documentation of results at depths both above and below 5 m b.s. (Christiansen et al., I, III, and IV). Special emphasis is therefore placed on results from this study in the following.

4.1 Direct documentation methods

Results of enhanced delivery can be documented both indirectly and directly. The indirect techniques most commonly utilized and valid reasons for doing so are described briefly in Christiansen et al (II). Direct documentation methods provide indisputable physical evidence of induced fracture characteristics and substance distribution. Direct documentation methods include augering, coring, and excavation, often coupled with geological characterization.

Table 4.1
Directly documented characteristics of fractures induced via fracturing in clay-type settings (modified from Christiansen et al., II and IV)

Characteristic	Hydraulic fracturing	Pneumatic fracturing
Targeted fracturing depth	Most commonly within 2-5 m b.s. (e.g. Roote, 2000); Extremes 1-24 m b.s.	
Number of directly documented fractures	~200 fractures (induced at ~20 sites)[A]	12 fractures (induced at 2 sites)[B]
Directly documented fractures at depths > 5 m b.s.	~20 fractures[C]	3 fractures (Christiansen et al., IV)
Fracture orientation and form	Hydraulic fractures induced at shallow depth (< 5 m b.s.) are typically horizontally-oriented (e.g. Murdoch and Slack, 2002)	Very few pneumatic fractures have been documented in available literature, and results are ambiguous (Markesic, 2000; Christiansen et al., I)
	Direct documentation of fractures induced at depths exceeding 5 m b.s. are highly variable – a tentative guideline for fracture propagation patterns is proposed in Section 3.3.3	
Distribution radius	4.5-21m omferred via indirect documentation methods (e.g. Roote, 2000); 1-3 m documented via direct methods in Danish clay tills[D]	1-17 m stated via indirect documentation methods (e.g. Schuring, 2002); 1-2 m (larger along isolated pathways) documented via direct methods in Danish clay till (Christiansen et al., I and IV)
	Note the distinction between distribution radius and radius of influence, as this has not always been observed in the literature.	
Fracture aperture	Commonly stated: 0.5-2 cm (e.g. USDoE, 1998); Surrogate value = surface uplift during fracturing =0.3-4.65 cm based on 16 shallow fracturing applications[E]; 0.1-5 cm indicated via direct documentation[F]	Commonly stated: 0.5-1 mm (e.g. USDoE, 1998); Values of surface uplift = 0.02-4.90 cm based on 4 studies[G]; 0.1-2 cm indicated via direct documentation (Christiansen et al., I)
	Surface uplift not expected to represent aperture adequately at depth (exceeding 5 m b.s.; Murdoch & Wilson, 1994; Christiansen et al., IV).	

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Characteristic	Hydraulic fracturing	Pneumatic fracturing
Thickness of induced reaction zones	Dependent on chosen *in situ* remediation treatment, see Section 3.2.1. Amendment-comparable tracers utilized in Christiansen et al. (IV) created diffusion zones up to 20 cm thick within 3-4 months of delivery.	
(Vertical) spacing of fractures	Typical norm 0.5-1 m (e.g. Bures, 1998 and 2010); 15 cm documented without merging at shallow depth (Murdoch et al., 1991)	Typical norm 0.5-1 m, due to alleged ability to create dense networks of fractures? (e.g. USDoE, 1998)

[A](Murdoch et al., 1991; Murdoch, 1995; Siegrist et al., 1999; Wong & Alfaro, 2001; Murdoch and Slack, 2002; Walsted et al., 2002; Bures et al., 2004; Westergaard, 2005; Blem et al., 2006; Butler-Vetyia et al., 2006; Murdoch et al., 2006; Jørgensen et al., 2007; Butler-Vetyia et al., 2008; Klint et al., 2008; Christiansen et al., IV)
[B](Markesic, 2000; Christiansen et al., I)
[C](Murdoch et al., 2002; Bures et al., 2004; Blem et al., 2006; Jørgensen et al., 2007; Christiansen et al., IV)
[D](Walsted et al., 2002; Westergaard, 2005; Blem et al., 2006; Jørgensen, 2007; Christiansen et al., IV)
[E](Murdoch et al., 1991; Frank & Barkley, 1995; Bures, 1998; Nilsson et al., 2000; Roote, 2000; Murdoch & Slack, 2002; Bures et al., 2003-4; Strong et al., 2004; Murdoch et. al., 200;)
[F](Murdoch et al., 1991; Siegrist et al., 1999; Murdoch and Slack, 2002; Murdoch et al., 2006; Klint et al., 2008, Christiansen et al., IV)
[G](Venkatraman, 1998; Roote, 2000; Palaia and Sprinkle, 2004; Strong et al., 2004)

To ensure that deliveries/induced fractures are recognizable, colored tracer and/or sand may be delivered. Excavation entails a great deal more site disruption than augering and coring, but is an unparalleled opportunity to collect empirical evidence of fracture/substance distribution and to perform geological site characterization. Augering provides only disturbed samples of the fractured subsurface, while coring balances sample integrity with minimal site disruption.

Environmental fracturing has not been systematically directly documented at depths exceeding 5 m b.s. (Christiansen et al., II). As stated earlier, direct-push delivery, which has only been used in low-permeability deposits in recent years, also lacks sufficient documentation of its capabilities here. This has been addressed by the research conducted for this PhD project (Christiansen et al. I, II, III, and IV). However, the results of these studies have highlighted the need for further research before the environmental fracturing technology can be considered reliable.

It is important to note that water sampling (indirect documentation), which is frequently used to monitor *in situ* remediation progress, can be alluringly positive. Often misleadingly so in low-permeability sediments, as water collection filters are generally placed in or collect water from high-permeability zones. Here, remediation is most likely more advanced than in the main sediment volume (the low-permeability matrix). Soil sampling results often provide a more accurate picture of remediation progress (Broholm et al., 2010).

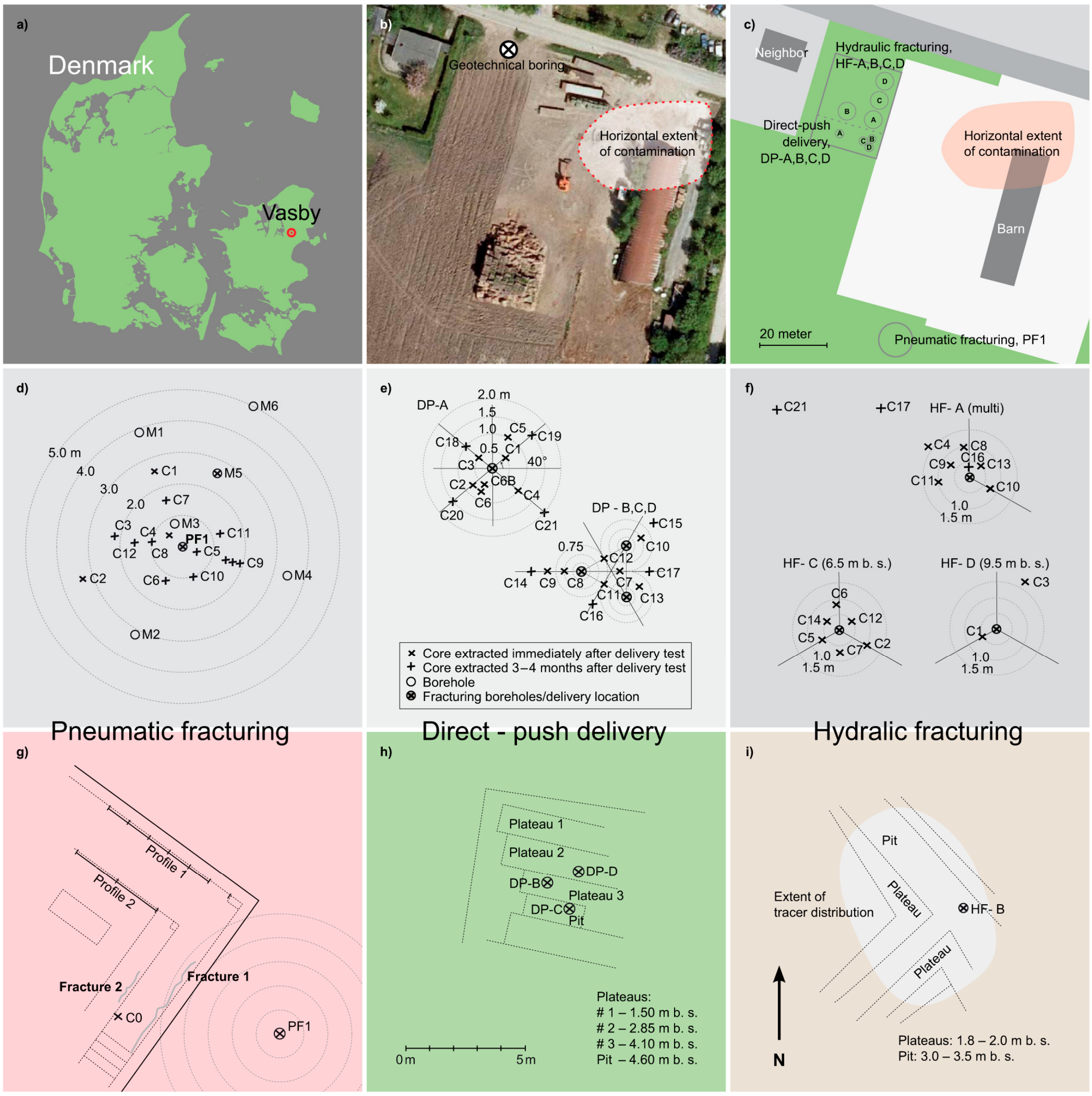

Figure 4.1: Overview of comprehensive field testing of the three enhanced delivery methods at a clay till site in Vasby, Denmark. (a) Location of Vasby in Denmark. (b) Schematic of field site including individual test plots and layout. (c) Aerial photo of field site. (d) Coring and augering locations around the pneumatic fracturing borehole. (e) Coring locations around the direct-push delivery spots. (f) Coring locations around the hydraulic fracturing boreholes. (g) Excavation conducted in the periphery of expected pneumatic fracturing influence zone. (h) Excavation conducted over cluster of direct-push delivery locations. (i) Excavation conducted over hydraulic fracture induced at 3 m b.s. From Christiansen et al. (IV).

4.2 Induced fractures

The induced fracture characteristics important to remediation design were mentioned in Section 3.1, and include achievable depths, orientations, forms, distribution radii, apertures, and spacing. Christiansen et al. (II) present directly documented induced fracture characteristics. This work is briefly summarized in the following Sections 4.2.1 – 4.2.4 and compared to directly documented direct-push deliveries undertaken by Christiansen et al. (III, IV).

Combining knowledge of fracture characteristics with knowledge of selected *in situ* remediation amendment characteristics can facilitate estimation of probable reaction zone thicknesses and mass removal rates at a given site. Potential reaction zones thicknesses have been indicated by the use of tracers (comparable to some *in situ* remediation amendments with regard to diffusivity/mobility) in Christiansen et al. (I, III, and IV), but tests with actual amendments were beyond the scope of these studies. It has therefore also been beyond the scope of this project to investigate probable mass removal rates, but observed trends to date are briefly reported (Section 4.3).

4.2.1 Depths, orientations and forms

The most common fracturing depth interval at clay-type sites is 2 – 5 m b.s. (Roote, 2000), and induced hydraulic fractures are typically flat-lying and elliptical (as the ideal form) at these depths (Murdoch, 1991; Murdoch and Slack, 2002; Murdoch et al., 2006). Approximately 20 hydraulic fractures emplaced at depths exceeding 5 m b.s. in clay-type sediments (at 6 sites) and directly documented via coring are found (Murdoch et al., 2002; Bures et al., 2004; Blem et al., 2006; Jørgensen et al., 2007; Christiansen et al., IV). Depths varied from 6-13.2 m b.s. and the orientations of these fractures were highly variable: 9 dipped gently upwards (<35°), 9 dipped more steeply upwards (36-60°), 9 had a near vertical orientation (>60°), 5 curved downwards, and 1 (Christiansen et al., IV) was not located, despite extraction of 7 soil cores surrounding the fracturing borehole. The dip of hydraulic fractures increased with depth at some sites, but flattened with depth at others due to varying geological and geotechnical conditions. While surface surveying during fracturing generally indicated relatively circular fracture extents, coring results indicated highly variable, erratic forms.

Three pneumatic fractures were induced at one site at depths exceeding 5 m b.s. (5.5-7.5 m b.s.), and directly documented (Christiansen et al., I). Their dips were difficult to determine based on core and augering observations alone, but a partial excavation of the site revealed vertically-oriented tracer-filled natural fractures. It is assumed that induced fractures had an initially (sub)horizontal propagation path but were short-circuited by vertical natural fractures. Tracer propagation appeared to have continued along the natural fractures upwards toward the surface (Christiansen et al., I). Similar results were observed for more shallowly-induced pneumatic fractures at a Canadian clay till site (Markesic, 2000). Sporadic fracture observations in cores indicated spoke-like, rather than circular, forms of the fractures (Christiansen et al., I and IV).

The results of closely-spaced deliveries at depths from 6-9.5 m b.s. in four direct-push delivery locations have been directly documented via coring at the Vasby clay till site. Direct documentation of delivery orientations and forms has not been conducted elsewhere. Most delivery points were visible in cores up to 1 m away from the delivery location, at the approximate depth of their delivery (Christiansen et al., III and IV).

4.2.2 Distribution radii

Indirectly documented values of induced fracture radius fall within a broad range from 0.9-21 m for hydraulic fracturing and 1-17 m for pneumatic fracturing (e.g. Roote, 2000; Schuring, 2002), and it is not always clear whether fracture radius or radius of influence (a term for hydraulic response) is reported. The latter, henceforth ROI, refers to the radius within which effects of fracturing can be measured, although physical evidence of fractures may not be observable. This parameter has commonly been measured due to the previous focus on fracturing-assisted remediation involving mass transfer methods. Directly documented induced fracture radii at depths exceeding 5 m b.s. are few.

Coring results from fracturing tests in Danish clay tills indicate hydraulic fracture distribution radii at depths exceeding 5 m b.s. on the order of 1-3 m (Westergaard, 2005; Jørgensen et al. 2007; Christiansen et al., IV), pneumatic fracture distribution radii of 1-2 m (larger along isolated pathways, Christiansen et al., I and IV), and direct-push distribution radii of ~1 m (Christiansen et al., III and IV).

4.2.3 Apertures

Measurement of surface uplift has generally been accepted and used as a proxy for induced fracture aperture for fractures that are broad relative to their depth (length = 3 x depth) (Murdoch & Wilson, 1994). Values for surface uplift during hydraulic fracturing range from 0.3-4.65 cm based on 16 studies conducted at 0-5 m b.s. (Bures, 1998; Bures et al., 2003-4; Frank & Barkley, 1995; Murdoch et al., 1991; Murdoch et al., 2006; Murdoch & Slack, 2002; Nilsson et al., 2000; Roote, 2000; Strong et al., 2004). Directly documented hydraulic fracture apertures and sand thicknesses fall within a similar range from 0.1-5 cm at depths until 5 m b.s. (Murdoch et al., 1991; Siegrist et al., 1999; Murdoch and Slack, 2002; Murdoch et al., 2006; Klint et al., 2008, Christiansen et al., IV). The apertures of individual hydraulic fractures were observed to vary significantly, both thickening and thinning intermittently from initiation point to tip, see example in Figure 4.2a. Christiansen et al. (IV) report fracture apertures of 0.1-0.5 cm for fractures emplaced at 6-7 m b.s., while the maximum corresponding surface uplift was 1.4 cm, indicating poorer correlation between surface uplift and fracture aperture, when fractures are emplaced at depths exceeding 5 m b.s.

Values for surface lift observed during pneumatic fracturing range from 0.02-4.90 cm based on 4 studies (Palaia and Sprinkle, 2004; Roote, 2000; Strong et al., 2004; USEPA, 1995; Venkatraman, 1998). The lower end of the range stems from a pneumatic fracturing application at 18-24 m b.s. (Strong et al., 2004; Palaia and Sprinkle, 2004), while the larger values (0.3 – 4.9 cm) stem from shallow applications. Directly documented pneumatic fracture apertures are only reported in Christiansen et al. (I and IV), and range from 0.1 to 2 cm (observed in cores and excavation) for fracturing depths of 4-8 m b.s. (see Figure 4.2b).

It was not possible to gauge apertures of the fractures induced via direct-push delivery at the Vasby clay till site in Denmark (Christiansen et al., III and IV), as the sorbing tracer rhodamine WT was not visible (under UV-light).

4.2.4 Reaction zone thicknesses

The mobile tracer fluorescein delivered during field tests at the Vasby site quickly diffused into the sediment creating tracer-affected zones up to 6 cm wide initially and up to 20 cm wide after 3 months (see Figure 4.2b). The tracer is comparable to diffusive *in situ* remediation amendments (e.g. permanganate, see Section 3.2.1), while the other fluorescent tracer employed, rhodamine WT, is strongly sorbing and thus more similar to ZVI (see Section 3.2.1). Clearly, reaction zones stemming from both large- and small-aperture fractures (i.e. hydraulic, pneumatic and direct-push) can reach significant widths within a short timeframe.

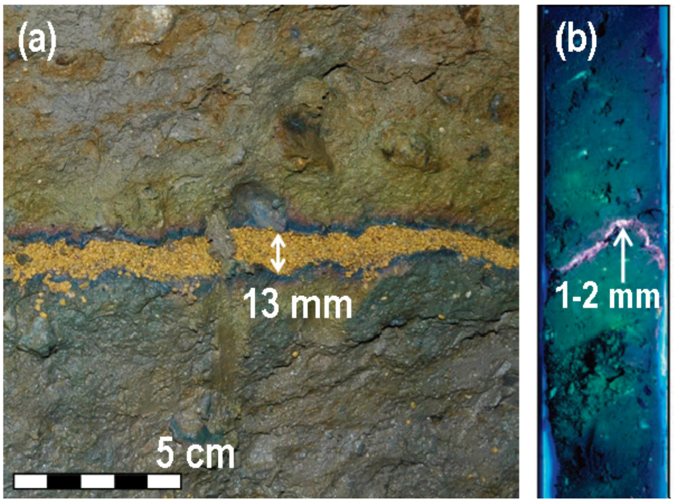

Figure 4.2: (a) Excavated hydraulic fracture induced at 3 m b.s. at the Danish Vasby clay till site. The fracture induced with yellow fracturing sand, and subsequently a tracer mixture which appeared purple in daylight conditions was injected into the fracture. In the figure the tracer mixture is seen to diffuse into the sediment surrounding the yellow sand-filled fracture. Note the variable fracture aperture/thickess. (b) Pneumatic fracture filled with fluorescent tracers (appearing pink and green) under UV-light at 5.75 m b.s. in a core. A sorbing and a mobile tracer have been delivered to the fracture (3-4 months prior to core collection). Fluorescein (the green) diffused significantly into the matrix, while Rhodamine WT stayed sorbed to fracture surfaces. From Christiansen et al. (II).

4.2.5 Spacing

As already stated in Section 3.2, closely-spaced fractures and deliveries of *in situ* remediation amendments are crucial to overcoming mass transport limitations imposed by diffusion at chloro-ethene-contaminated low-permeability sites (Chambon et al., 2010). Hydraulic fractures were successfully induced with a mutual distance of as little as 15 cm without causing fractures to merge at depths of 1-3 m b.s. by Murdoch et al. (1991). Despite these positive results, the norm is to emplace fractures with mutual spacing of 0.5-1 m. Reasons for this are not specifically reported, but they are likely to be budget constraints, underestimation of diffusion limitations on remediation timeframes (and likely rebound of contaminant concentrations after initial decreases), and/or unresolved remediation goals/success criteria. Christiansen et al. (IV) therefore recently attempted to induce 4 hydraulic fractures in one borehole in a clay till, each 25 centimeters apart over a depth interval of 6.25-7 m b.s. The attempt was not successful: One of the fractures was not observed, despite extensive coring, while the others are seen to merge short distances from the borehole (0.25-0.75 m) and incline quite steeply towards the surface (30-60°). Attempts at close spacing of pneumatic fractures have not been reported, likely due to its alleged ability to create networks of small-aperture fractures (e.g. USDoE, 1998). Direct-push delivery, on the other hand, is generally used to deliver amendments from closely-spaced delivery points (10-25 cm), and Christiansen et al. (III and IV) have documented that this can be done successfully.

4.2.6 Summary of Vasby field test results

As stated earlier, the Vasby field study of pneumatic fracturing, direct-push delivery, and hydraulic fracturing is the only field study comparing all three enhanced delivery methods under similar conditions. In addition, values for parameters that are required to design remediation projects were obtained. Results are summarized in Table 4.2 below.

Table 4.2

Summary of values for important reaction zone parameters indicated by the Vasby enhanced delivery pilot tests. From Christiansen et al. (IV).

Reaction zone parameter	Value indicated for			
	Pneumatic fracturing	**Direct-push delivery**	**Hydraulic fracturing, 3 m b.s.**	**Hydraulic fracturing, >6 m b.s.**
Thickness – sorptive amendment	0.1-0.5 cm	0.1-0.5 cm ?	0.1-2 cm	
Thickness – mobile amendment (3-4 months after delivery)	10 cm	10-20 cm	Same order of magnitude as the other methods?	
Spacing (lower limit)	< 1 m ?	10 cm	?	
Orientation	Subhorizontal but prone to (vertical) diversion in basal clay till?	Subhorizontal	Sub-horizontal	?
Form	Spoke-like?	Circular	Elliptical (off-center from borehole)	?
Distribution radius	2 m	1 m	3-3.5 m	?

4.3 Enhanced *in situ* mass removal

For remediation efforts assisted by environmental fracturing in clay-type settings, the most frequently targeted contaminants are chloroethenes (e.g.McKay et al., 1989; Roote, 2000; Butler-Vetyia et al., 2008). The most frequently chosen *in situ* remediation technologies have been the mass transfer technologies SVE and Dual Phase Extraction (DPE) (e.g. D'Astous et al., 1989; Roote, 2000; Doesburg, 2008). However, *in situ* mass removal technologies are increasingly used (e.g. Butler-Vetyia et al., 2008a,b; Chen et al., 2008). Quantitative figures for (chloroethene) contaminant mass removal vary from 50-100% (e.g. USEPA, 2003; Strong et al., 2004). Generally, a 90% reduction or greater in at least part of the remediation area is reported. In absolute figures, residual concentrations vary from the non-detectable to 200 ppm (mg/L). However, due to the lack of detail in the reporting of many remediation projects, it is unknown how results are derived (water vs. soil sampling, etc.) and hence to what extent results are reliable or even comparable. Initial results of remediation efforts assisted by environmental fracturing are not expected to be lasting: slightly longer-term measurements of mass removal rates often display falling rates and contaminant rebound (e.g. USEPA, 1995; Martin et al., 2002; Strong et al., 2004). This is attributed to diffusion limitations not fully overcome by the fracturing (Chapman & Parker, 2005), which is typically conducted with large spacing of fracturing levels (see Section 4.2.5).

Direct-push delivery applications at clay till sites in Denmark for enhancement of *in situ* mass removal via ERD have quantitative results similar to the above. Significant reductions in chloroethene concentrations have been achieved (Kjærsgaard, 2006a,b; Tsitonaki and Broholm, 2010), but presently, 3 years after initial deliveries, new amendment deliveries are required to prevent remediation stagnation (and contaminant mass rebounds). Interestingly, deliveries in these applications have been made with small spacing (25 cm).

4.4 Findings for documentation of enhanced delivery

- A literature review has revealed that directly documented characteristics of fractures induced via enhanced delivery are sparse at depths exceeding 5 m b.s. Findings are summarized in Table 4.1.
- Directly documented field tests with focus on geological and geotechnical site characterization are required to further the state of knowledge regarding orientations and forms of fractures induced by the enhanced delivery methods.

- Closely-spaced, subhorizontal substance deliveries can be achieved robustly with direct-push delivery in clay till.
- Preliminary values for parameters that are crucial in remediation design have been determined from the comparative field tests of enhanced delivery. Results are summarized in Table 4.2.

5 Discussion of enhanced delivery methods

The previous sections have described important geological and geotechnical aspects of low-permeability clay tills, methods for enhanced delivery of *in situ* remediation amendments in clay till, and documented results of these methods to date. Thus, it is clear why methods for enhanced delivery of *in situ* remediation amendments in clay till are a topic for debate and research.

The aim of this PhD project has been to shed light on the enhanced delivery methods, what can be achieved by their use, and parameters that are crucial for enhanced *in situ* remediation design. These goals have been addressed via a set of pilot tests for pneumatic fracturing, hydraulic fracturing, and direct-push delivery. Results of these comparative pilot tests of the enhanced delivery methods have been highlighted throughout the thesis and are thoroughly reported in the appendices (Christiansen et al., I, II, III, and IV). The tests were conducted at the same site, utilizing the same tracers and direct documentation methods. Furthermore, the test site was chosen based on its geological composition, which is thought to be representative of many contaminated sites in Denmark, and potentially worldwide. Whether this strategy has provided results that are mutually comparable and outwardly representative is discussed in the following. Based on this, the present status of the enhanced delivery methods is also discussed.

5.1 Comparability of the Vasby pilot tests

To facilitate comparison of the results of the pilot tests of pneumatic, hydraulic, and direct-push delivery conducted at the Vasby site, as many elements as possible were kept identical: site, season, tracers, and documentation methods. Elements that were not kept identical were: initiation and propagation pressures, introduced tracer amount per delivery point, introduced rhodamine WT concentrations, and number of delivery points and locations per test.

5.1.1 Variations in pressure and delivery volumes

The reason for inititation and propagation pressure variation was method dependency. The pressures required to initiate and propagate fractures were chosen by experienced technicians in each test and varied from 1.38-8.62 bar for pneumatic fracturing, 0.75-5.25 bar for hydraulic fracturing, to 19-30 bar for direct-push delivery[2] (Christiansen et al., IV). Similarly, each method is suited for introduction of varying amounts of substance per delivery point. For hydraulic fracturing, the amount of tracer introduced (250 L per fracture) was based on the expected approximate pore volume of emplaced fractures. The expected volume was calculated by assuming circular fractures with a radius of 3 m, aperture/thickness of 2 cm, and porosity of 40% (Christiansen et al., IV). For pneumatic fracturing, it was assumed that it would be possible to atomize 300-400 L of liquid to flow with the nitrogen gas within the typical short duration of pneumatic fracture propagation per fracturing interval. However, only 50 L of tracer mixture were introduced at every fracturing level during the Vasby pilot test in order to ensure recovery/observation of the tracers without injecting unnecessarily large tracer amounts into the subsurface (Christiansen et al., I). A maximum possible delivery amount was not estimated for direct-push delivery, but previous full-scale tests at clay till sites involving delivery volumes of 3-8 L per delivery point have been successful (Kjærsgaard, 2006a and b). Thus, a delivery volume of 10 L per delivery point was chosen for the Vasby pilot test of this method (Christiansen et al., III).

As the pressure ranges applied during the pneumatic fracturing and direct-push delivery tests actually overlap significantly, pressure variations are not believed to be responsible for the variations in tracer distribution observed for these two pilot tests. The introduction of smaller tracer volumes per delivery point during direct-push delivery may, however, be responsible for the main difference in tracer distribution characteristics between these two methods, for which observations otherwise are quite similar. Tracer distribution radius was ~1 m for direct-push delivery and ~2 m for pneumatic fracturing. Thus, it is possible that the delivery of a larger volume of tracer during direct-push delivery would have resulted in a larger tracer distribution radius.

During the hydraulic fracturing test, sand-filled fractures were emplaced first, and subsequently, tracer was injected into these fractures. Thus, the radius of tracer distribution was given by the achieved fracture radius, i.e. the injected tracer volume had no bearing on achieved tracer distribution radius. Furthermore, since lower pressures were generally applied during the hydraulic fracturing test (than the other delivery tests) and a larger tracer distribution/fracture radius was still achieved (at 3 m b.s.), variations in applied pressures are again not assessed

[2] Up to 19 bars were back.pressure in the probe, so direct-push delivery pressures were effectively somewhat lower.

to be responsible for the variations in tracer distribution observed for this pilot test compared to the others.

5.1.2 Variation of tracer concentration

The resulting concentration of rhodamine WT in the tracer mixture utilized in all three pilot tests was adjusted from 10 000 mg/L during the pneumatic fracturing test to 2 000 mg/L during the direct-push and hydraulic fracturing tests. This adjustment was made because rhodamine WT concentrations were an approximate factor of 5 higher than correspondingly measured fluorescein concentrations in soil samples from cores extracted 4 months after the pneumatic fracturing test (Christiansen et al., I; see Figure 3.3a). I.e. it was assessed that the use of a smaller amount of rhodamine WT (20%) would still ensure its observation in cores/fluorometer analyses, while limiting the amount of tracer introduced to the sediment.

This reduction in rhodamine WT concentration had one significant disadvantage. In cores collected after the pneumatic fracturing test, it was possible to discern the actual paths and approximate apertures of observed tracer-filled fractures by means of a clear magenta coloration of the fractures under UV-light by rhodamine WT (which sorbed to the fracture walls, see Figure 3.3a). This was not possible after the direct-push and hydraulic fracturing tests, as concentrations of the tracer were not high enough to generate the magenta coloration of fracture and tracer paths under UV-light (Figure 3.3b). Thicker bands of fluorescein-affected sediment (surrounding the actually induced fractures) were clearly observable during the direct-push delivery test, and hydraulic fractures were observable via the emplaced epoxy-coated sand (and surrounding fluorescein 'halo'), but it is not possible to say whether smaller (micro-)fractures were induced outwards from the main sand-filled fractures based on tracer observations.

5.1.3 Variation in number of delivery points and locations

The final variation between the enhanced delivery tests conducted at the Vasby site was the number of delivery points and locations of each test. This was again a matter of method dependency. Generally, the tests had the objectives of evaluating the distribution radius of the methods and showing whether closely-spaced horizontal fractures/substance deliveries could be achieved in both the weathered (0-5 m b.s.) and unweathered horizon (>5 m b.s.) of a clay till (Christiansen et al., IV). Thus, pneumatic fracturing was carried out in one location, as this type of fracturing was expected to have a relatively large distribution radius (5 m) and be suited for multiple fracturing levels in one location. The fracturing levels were widely spaced (8, 7, 6, 5, and 4 m b.s.) because pneumatic fracturing was furthermore expected to be able to induce dense networks of small-aperture fractures outwards from each fracturing level, making closely-spaced fracturing levels superfluous (Christiansen et al., I). Direct-push delivery was carried out in four locations due to the expectation of a small distribution radius (1 m) based on previous experiences with the method (Kjærsgaard, 2006a and b; Tsitonaki et al., 2009), and, hence, the desire to evaluate both the distribution radius for a single delivery location and the areal coverage for a cluster of (three) delivery locations. Delivery points were closely spaced (25 and 10 cm) in all delivery locations in compliance with the second objective of the tests (Christiansen et al., III). Hydraulic fracturing was also conducted in four locations, as multiple fractures are not usually emplaced in the same borehole. Three locations were thus fractured at one depth each, 3, 6.5, and 9.5 m b.s., respectively, to evaluate distribution radius, form and orientation at three different, relevant depths. At the fourth location, multiple, closely-spaced fractures (25 cm) were attempted emplaced at 6.25-7 m b.s. in one borehole using hydraulic fracturing (Christiansen et al., IV).

Thus, the variation in number of delivery points and locations for each of the enhanced delivery method tests at the Vasby site was necessary to fulfill the overall objectives of each individual test and the project as a whole. Hence, this variation is viewed as a feature that has enhanced, rather than reduced, comparability of the test results.

5.1.4 Evaluation of comparability

In conclusion, the only variation between the enhanced delivery tests conducted at the Vasby site that may have had a significant impact on comparability, is the use of a small delivery volume per delivery point during direct-push delivery. This may have resulted in a smaller distribution radius than could otherwise have been achieved using larger delivery volumes. However, since it has been demonstrated that direct-push delivery has many other merits (Christiansen et al., III and IV), this uncertainty is not critical. The variations in initiation and propagation pressures, amount of tracer introduced per delivery point, rhodamine WT concentrations, and number of delivery points and locations per test have thus not compromised the comparability of the test results achieved at the Vasby site. Hence, a basis for credible comparison of the three methods has been provided. Christiansen et al. (IV) conclude that direct-push delivery is presently, based on available documentation and a cost survey of the enhanced delivery methods, the most robust and cost-effective method of the three to enhance delivery of *in situ* remediation amendments in Danish clay till.

5.2 Representativity of Vasby results

The Vasby site is, based on extensive geological characterization work carried out in three excavations at the site, geologically representative of many contaminated sites in Denmark (Christiansen et al., I and IV). However, it is not necessarily geotechnically representative (see Section 3). While geological and geotechnical features important to assessing the fracturing-suitability of a site may correspond (e.g. subglacial deposition and overconsolidation) they may also be dissimilar.

Values for consolidation state cannot be generalized in clay tills. Measurements are necessary at each proposed fracturing site. However, it is evident from Table 3.1 that this has not been the norm to date. The limited data that are available suffice to illustrate the importance of geotechnical testing at the individual site. OCR and K_0 values derived at the Næstved, Haslev, and Vasby site suggest that the Næstved-till is overconsolidated, the Vasby-till normally to slightly overconsolidated, and the Haslev-till slightly underconsolidated, despite similar (sub)glacial origin. I.e. processes during or after deposition have inhibited consolidation at the Vasby and Haslev sites, and fractures have propagated accordingly.

The consolidation state of other low-permeability sediments (glacial and non-glacial) is equally difficult to generalize. For example, K_0 values both greater than 1 and less than 1 may be encountered in recent fluvial sediments depending on the influence of wetting and drying cycles in the given sediment. In contrast, older fluvial sediments may be overconsolidated due to burial, consolidation and, subsequently, unloading due to uplift. While these processes are to some extent revealed via study of local and regional geological history and/or physical geological characterization, chances are that one or more significant events may be overlooked or untraceable (Sladen and Wrigley, 1983).

Therefore, geological characterization (determining presence of natural fractures, sand features, and stratigraphic layering) at a proposed enhanced delivery site, coupled with geotechnical tests (anisotropically consolidated, undrained triaxial tests; Christensen et al., 1992) should never be forgone if any degree of certainty with regards to potential fracturing success is desired.

5.3 Status of enhanced delivery method capabilities

The literature review (Christiansen et al., II) has indicated that many unknowns persist with regard to the remediation enhancement capabilities of environmental fracturing. The role of slow diffusion in low-permeability matrices has often been underestimated or neglected, i.e. close fracture spacing has not been a priority/demand. Focus has been placed on short-term fluid extraction enhancements (e.g. increases in borehole radius of influence and yield post-fracturing). One could argue that the previous focus on mass transfer techniques, such as SVE and DPE (e.g. Roote, 2000), warranted this focus, but decreasing mass extraction rates and rebounds in contaminant concentrations over time suggest the necessity of diffusion limitation considerations also in this context, and, hence, the inducement of closely-spaced fractures.

Christiansen et al. (I and IV) have worked to convey the importance of understanding the dependency of the enhanced delivery technologies on the geological and geotechnical features of a given sediment, and provide concrete values for the parameters essential to fracturing-assisted remediation design – in the unweathered horizon of a clay till (>5 m b.s.). Based on this work, it seems that especially hydraulic fracturing is very sensitive to *in situ* consolidation conditions, whereas pneumatic fracturing and direct-push delivery are less, but not insignificantly so. A near-horizontal fracture was achieved only in the weathered horizon with hydraulic fracturing. All tests in the unweathered horizon, including the attempt to emplace multiple closely-spaced fractures in one borehole, were unsuccessful. Horizontal fractures were observed in the unweathered horizon at the pneumatic fracturing location. However, a propensity for fracture diversion and cessation in natural high-permeability features (e.g. fractures and sand lenses) is expected for pneumatic fracturing. Furthermore, close spacing was not achieved in the unweathered horizon. Closer spacing of pneumatic fracturing depths may change this. Horizontal, closely-spaced distribution was achieved consistently throughout the direct-push delivery test. Increased connectivity to vertical fractures was not assessed as having a negative effect on lateral distribution radius (Christiansen et al., III).

5.3.1 Potential further development of enhanced delivery methods

Whether hydraulic fracturing would fare better in the unweathered horizon of a (clay till) sediment with different geotechnical features than those encountered at the Vasby site is difficult to assess. However, it is not believed that alterations to any technical or procedural aspects of the hydraulic fracturing method, as it was implemented at the Vasby site, would significantly alter the results achieved here. Pneumatic fracturing may, on the other hand, achieve better results (i.e. closely-spaced fractures) if the fracturing assembly was redesigned to permit a much closer fracturing interval: The utilized nozzle and packer system was long (about 4.65 m) and only permitted a fracturing interval of about 0.9 m. Direct-push delivery achieved good field results at the Vasby site. A trial involving evaluation of distribution radius of larger delivery volumes per delivery point would, however, be beneficial.

Furthermore, potential problems with upscaling of pilot tests to full-scale tests must always be anticipated based on the expansion of affected soil volume and consequently altered system boundary conditions. This is exemplified by results obtained by Tsitonaki et al. (2009). During their pilot testing of direct-push delivery for enhanced delivery of persulfate for ISCO in a Danish clay till, a total of 250 L of persulfate were delivered without problems from 10 delivery points within a soil volume of approximately 8 m^3 (~30L/m^3). Upon full-scale application, 1610 deliveries of each 25 L were planned within a soil volume of 160 m^3 (~250 L/m^3). However, only one third of the planned deliveries were carried out (~80 L/m^3) due to extensive venting of persulfate at the surface.

5.3.2 Efficacy of the enhanced delivery methods

Based on the North American experiences with environmental fracturing briefly described in Section 4.3, shortcomings of fracturing-assisted remediation schemes are attributed to inadequate fracture spacing. It is noted, however, that Danish applications of direct-push delivery with closely-spaced delivery points also experience contaminant rebound. I.e. initial amendment delivery volumes have not been able to completely remediate the targeted contaminant mass. This indicates that the volume of delivered amendments was insufficient. In actuality, however, it is typical remediation strategy to deliver necessary amendments via several delivery rounds over time (Kjærgaard, 2006a,b; Tsitonaki et al., 2010). This is done in recognition of limitations on amendment efficiency due to suboptimal *in situ* conditions, and to facilitate better distribution and utilization of delivered amendments over time.

Thus, indications are that the enhanced delivery methods can serve to adequately enhance complete *in situ* mass removal setups if closely-spaced, horizontal fractures can be induced. Incremental delivery may be deemed advantageous in some cases, and free of upscaling issues. However, close spacing of hydraulic and pneumatic fractures at depths exceeding 5 m b.s. has yet to be documented. An effective test of this could be facilitated by performing fracturing at a site which has been characterized as having geotechnical characteristics at depths greater than 5 m b.s. similar to sites that have provided ideal fractures at shallow depths.

6 Conclusions

This PhD project has studied the technologies available for enhancing delivery of *in situ* remediation amendments in clay till. These technologies are pneumatic fracturing, hydraulic fracturing, and direct-push delivery. The following key findings have been made:

- A literature review has revealed that systematic correlation of geological and geotechnical properties of fracturing/enhanced delivery sites with resulting induced fracture characteristics is lacking.

- Thorough geological characterization has revealed that the Vasby field site is a normally consolidated, extensively naturally fractured basal clay till (type B).

- Pneumatic fracturing is capable of distributing amendment-comparable tracers in subhorizontally-oriented, spoke-like fractures with a distribution radius of up to 2 m at depths of 4-8 m b.s. in the Vasby till. Fractures are prone to short-circuiting in natural vertical fractures.

- Direct-push delivery is capable of distributing amendment-comparable tracers in closely-spaced, subhorizontally-oriented fractures with a distribution radius of at least 1 m at depths of 2.5-9.5 m b.s. in the Vasby till. The delivery method performs robustly in all (4) delivery locations.

- Hydraulic fracturing is capable of distributing amendment-comparable tracers in a distinct subhorizontal fracture with a fracture radius of up to 3.5 m at 3 m b.s. in the Vasby till. Distribution radius exceeds fracture radius. Fracturing at larger depths did not succeed in producing similar results.

- Based on literature study and field tests, a tentative guideline for fracture propagation patterns is proposed (Section 3.3.3).

7 Further research

The results of the enhanced delivery pilot field tests conducted for this PhD project stand alone, as direct documentation of fractures induced at depths exceeding 5 m b.s. is very sparse. Furthermore, all three enhanced delivery methods have never before been tested at the same site. Suggestions for further research are listed below:

- Attempts to document induced fracture apertures were not successful for all three methods, and should be investigated further, as should the importance of fracture aperture on substance distribution. Indications from the pilot tests are that fracture aperture is not influential, given the achieved tracer-affected zones from both small- and large-aperture fractures. The thicknesses of potential reaction zones were investigated using only tracers comparable to *in situ* remediation amendments. Further testing using actual *in situ* remediation amendments should be undertaken to ensure that their behavior is indeed similar.
- To further develop the use and reliability of the enhanced delivery methods, field tests ought to be carried out and directly documented via excavation and coring at low-permeability sites displaying significant geological and geotechnical characteristics both similar to and different from the Vasby site. Tests at similar sites would serve to validate or disprove the representativity of the Vasby test results, while tests at differing sites would serve to validate or disprove the proposed guideline for fracture propagation patterns.
- Investigation of the direct-push delivery method and feasible delivery volumes per delivery point should be undertaken. Presently, it is unclear how large a volume can be delivered per delivery point without complications, and whether delivery of larger volumes per delivery point will increase the distribution radius. Naturally, feasible delivery volumes will vary depending upon the sediments and chosen *in situ* remediation amendments, but it is expected that guidelines may be formed.
- The diffusive capabilities of *in situ* remediation amendments and the formation of micro-fractures outwards from induced fractures should be investigated. Significant diffusivity (mobility) of amendments shortens the distance that contaminants must travel to come into contact with them in the subsurface, while the formation of micro-fractures may facilitate placement of those amendments that do not diffuse on account of sorption or size, further into low-permeability matrices. When these processes are better understood, the need for closely-spaced fractures may be readdressed.

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9 Appendices

I Christiansen, C.M., Riis, C., Christensen, S.B., Broholm, M.M., Christensen, A.G., Klint, K.E.S., Wood, J.S.A., Bauer-Gottwein, P., and Bjerg, P.L. (2008): Characterization and Quantification of Pneumatic Fracturing Effects at a Clay Till Site. *Environmental Science & Technology* 42 (2): 470-576. DOI: 10.1021/es071294s.

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The papers are not included in this www-version, but can be obtained from the Library at DTU Environment. Contact library@env.dtu.dk or Department of Environmental Engineering, Technical University of Denmark, Miljoevej, Building 113, DK-2000 Kgs. Lyngby, Denmark.

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